Accelerator-Based Conversion of Weapons-Grade Plutonium

by

Robert A. Taylor

Submitted to the Department of Mechanical Engineering
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Abstract

Nuclear confrontation between the United States and the former Soviet Union is less likely now that political tensions have eased and a large portion of their nuclear arsenals are being dismantled. However, negative ramifications of dismantlement have raised the specter of nuclear proliferation in a new and dangerous arena: nuclear terrorism. As a result of warhead dismantlement, stockpiles of weapons-grade plutonium (WGPu) and highly-enriched uranium (HEU), which form the heart of a nuclear weapon, are steadily increasing in both the U.S. and Russia. There is concern, particularly for the Russian inventories, that adequate controls and facilities are not in place to handle the expected levels of plutonium and the even larger amounts of uranium from dismantled weapons. Coupled with the pressures of a volatile political and economic environment, the lack of secure storage capacity increases the risk of nuclear materials being diverted into the hands of non-nuclear states and terrorist groups. Recognizing this danger, Russia has solicited financial assistance from the U.S. for the construction of a secure underground facility for interim storage of the WGPu and HEU. It is expected that a similar facility (or facilities) will be built in the United States.

Plutonium disposition poses a particularly difficult problem. Unlike HEU, which can be mixed with natural uranium - thus rendering it unusable for nuclear weapons - and burned in existing commercial reactors, plutonium-fueled reactors are not well developed. Moreover, plutonium cannot be denatured; any mixture of plutonium can be made into a nuclear weapon. A variety of plutonium processing options have been proposed to render the WGPu more "proliferation resistant" for possible reuse in nuclear weapons, ranging from mixing WGPu with radioactive poisons to complete destruction through fissioning. However, a permanent plutonium disposal option has not been chosen by either country.

This thesis evaluates the accelerator-based fission reactor concept (ABC) that has been proposed to completely eliminate the WGPu through fissioning. The ABC concept constitutes an alternative to current solid-fuel reactors, and is a recast of early liquid-fueled reactor concepts, with one exception: the ABC will operate subcritically by means of a powerful linear accelerator that injects an exogenous source of neutrons into the core to maintain constant power level.

Based on the existing policy objectives of the U.S. and Russia, a general framework is developed to evaluate the ABC system as a plutonium disposition technology. In summary, this analysis shows that the ABC is a feasible concept, though its technical immaturity makes it less attractive than concepts that are based on near-term technologies. However, reactors with high plutonium consumption rates, such as the ABC, may become attractive, or even necessary, if the analysis is expanded beyond weapons-grade plutonium to include the larger stockpiles of reactor-grade plutonium.

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Chapter 1

Introduction

The most recent arms control agreements between the United States and the former Soviet Union, START I in 1991 and START II in 1992, have set into motion a process that will reduce the present stock of strategic and tactical nuclear weapons by approximately 65 percent [vMF$^+$, 1993]. It is estimated that Russia will recover approximately 90 metric tons of plutonium and 450 metrics tons of highly enriched uranium (HEU) from this process [BDF$^+$, 1992]. In the United States, the plutonium stock is expected to grow to a level of about 45 metric tons of weapons-grade plutonium (WGPu). Quantities of HEU could be much larger.

Although a relaxation of political tension between the superpowers has allowed them to reach such agreements, and has reduced the risk of nuclear confrontation, the large stocks of weapons material emerging from dismantlement raises a new and serious nuclear proliferation issue: how to store and dispose of the stocks of WGPu and HEU such that the risk of diversion into the hands of terrorists, other sub-national groups, as well as states with nuclear ambitions, is minimized. This question is particularly critical in Russia where the highly unstable economic and political conditions increases the chance of a successful diversion of WGPu and HEU. For the HEU, there is a straightforward technical fix; denature to low enrichment (2% to 4%) for use in a nuclear power reactor. Weapons-grade plutonium, on the other hand, cannot be denatured in this fashion, which makes the issue of its disposal, both technically and politically, a very difficult proposition. Here, we concentrate on plutonium disposition.

Isotope	*Weapons Grade Pu*	*Reactor Grade Pu*
Pu238	0.0%	1.4 %
Pu239	94%	54.5%
Pu240	5.8%	25.5%
Pu241	0.2%	13.5%
Pu242	0.0%	5.1%
critical mass	11 kg	13 kg

Table 1.1: Plutonium isotopes concentration in Weapons-grade and Reactor-grade plutonium (Trapp, 1993 and MLD, 1981)

The Threat

If WGPu were acquired by a terrorist group, it would be possible for them to produce a crude nuclear weapon, disperse the highly toxic material into the environment as a poison, or use it as a tool for nuclear blackmail. States could do the same, but the greatest danger is that they would fabricate the material into high yield nuclear bombs. However, the main difference between the risk posed by national entities versus terrorist groups is that states have a strong incentive, as members of the international community, to restrict the use of nuclear weapons even if available, terrorists do not.

The amount of plutonium needed for a particular weapon yield depends, not only on the isotopic quality of the plutonium, but also on the design, which in turn depends on the skills of the weapons designers and fabricators. The first nuclear explosive, the Trinity implosion bomb, contained about six kgs of WGPu. Table 1.1 shows the difference in isotopic concentration between weapons-grade and reactor-grade plutonium (RGPu). Weapons-grade data and critical mass values for both plutonium types is provided by Trapp [Trapp, 1993]. The isotopic concentration for the reactor-grade plutonium is taken from Malbrain *et al.* [MLD, 1981]. Weapons-grade plutonium consists mainly of Pu^{239} with only a few percent of Pu^{240} and other higher plutonium isotopes. As the concentration of the even isotopes, particularly Pu^{240}, increases, the "quality" of the plutonium, in terms of bomb design, decreases. Thus, RGPu is much less attractive for weapons, particularly for sub-national groups who lack the skill to compensate for the less favorable isotopics with a more advanced design.

In addition to having the required plutonium, producing an operational warhead requires a significant technical base. Mark [Mark, 1990] notes that one of the main barriers faced

by a would-be nuclear terrorist is the required competence in technical specialties such as shock hydrodynamics, critical assemblies, chemistry, metallurgy, machining, and electrical circuits. However, the technical gap between acquiring a sufficient amount of WGPu (or RGPu) and producing an operational nuclear device may be closing. Jenkins [LA, 1986] notes that:

> Although the ease with which a bomb could be made has probably been exaggerated in the popular press, the notion that some group outside the government programs can design and build a crude nuclear bomb is certainly more plausible now than is was 30 or 40 years ago... [T]he requisite technical knowledge has since gradually come into public domain.

1.1 Traditional Nuclear Proliferation Concerns

Until now, there have been two nuclear proliferation scenarios which have been of most concern to the international community: (1) construction of dedicated nuclear weapons productions facilities by non-nuclear weapons states including: reactors for producing plutonium, enrichment plants for producing HEU, and reprocessing facilities for separating plutonium from reactor fuel discharge; and (2), the diversion of RGPu, from the commercial fuel cycle, into a bomb production program. Thus, the line between using nuclear technology for peaceful and military purposes is difficult to draw, and it makes the job of preventing the proliferation of nuclear weapons a very complex proposition.

Albright cites North Korea as an example of the first case [Albright, 1993]. He reports that North Korea constructed a secret 5MWe nuclear plant and corresponding plutonium separations facility, in the late 1980s, with enough capacity to produce plutonium for one nuclear device per year. Although North Korea has admitted to the International Atomic Energy Agency (IAEA) that small quantities of plutonium have been separated, the number of bombs produced, if any, have not been verified. Because of the attention and pressure imposed by the international community on North Korea, they have threatened to withdraw as a non-weapons state signatory to the Non-Proliferation Treaty of 1968. North Korea claims this plant is a civilian facility, but the world suspects it to be a clandestine weapons plant.

Proliferation concerns involving the commercial nuclear fuel cycle are primarily centered on the possibility of diverting RGPu in mixed oxide (MOX) fuel. MOX fuel is a mixture

of RGPu in oxide form, and uranium oxide. Some fear that a nuclear fuel economy, which includes separating and storing plutonium for fuel fabrication, will increase the chance that the material will end up in the hands of those desiring to produce bombs.

1.2 The New Proliferation Threat

The acquisition of nuclear weapons by terrorist groups has become a new focus of proliferation concern by the international community. In the past, nuclear weapons capability has been exclusive to those with a sufficient industrial and technical base to produce them. Now, the concern is that Russia's political and economic instability, coupled with their growing WGPu stockpiles, open up a second, and substantially easier, avenue to acquire bomb material, thus, making it accessible to sub-national groups such as terrorist organizations. Victor Alessi, Director of Arms Control and Nonproliferation of the United States Department of Energy, agrees with this point and further notes that Russia and the other republics simply do not have the technology for transporting or storing the weapons material [Perlman, 1992], which augments the dangerous situation.

Looking at Figure 1-1, two possible diversion scenarios exist in Russia's military production sector. First, diversion may occur while the plutonium is being transferred to, or while in temporary storage. This case is particularly serious because the plutonium is still in weapons-grade form and is readily usable in a nuclear device. In the second case, diversion takes place after the plutonium has undergone processing to make it more proliferation-resistant. The likelihood of this scenario depends on the process chosen which, in turn, determines its isotopic composition, physical and chemical form, and strength of radioactive barriers.

To date, rumors have persisted that small quantities of fissile material [MR, 1992], as well as nuclear warheads, have been diverted from the former Soviet Union. Although there are no confirmed reports that warheads or materials have been diverted, von Hippel *et al.* [vMF$^+$, 1993] urge that "it is imperative that arrangements be agreed on that will allow monitoring and assistance from the West."

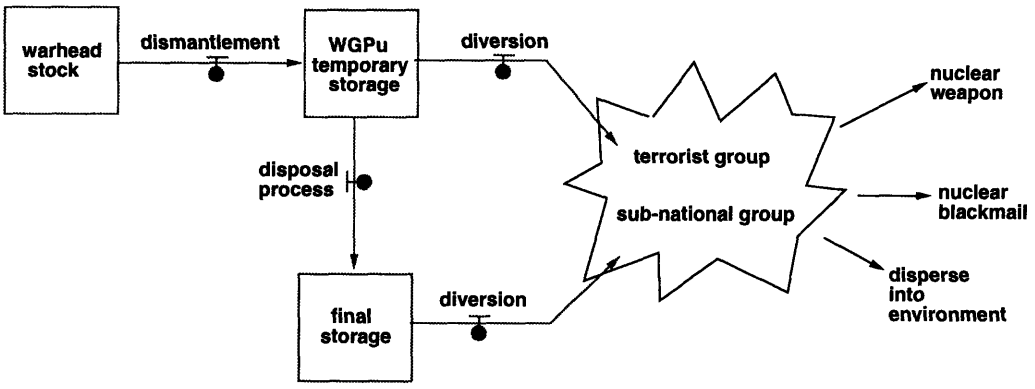

Figure 1-1: New proliferation threat from nuclear dismantlement

1.3 Plutonium Disposition Options

Interim Plutonium Disposal

The United States has had little time to react to the dramatic changes taking place in the former Soviet Union, particularly with regard to the dangers posed by the growing stocks of plutonium. Thus far, the main concern has been to ensure that all material is accounted for and stored under conditions that can withstand the social and economic pressures in Russia.

The incentive to divert the WGPu, HEU, and other specialized materials (e.g. ^{3}H and Be, and even nuclear skills) comes from within and outside of Russia, and is increasing as the Russian economy worsens. From within, there is a realization that these materials and skills could demand a high price to those desiring nuclear capability. On the outside, the price of nuclear capability has suddenly become much cheaper, as the research and development costs, the infrastructure requirements, and time needed to attain this capability can now be bypassed completely. Wald concludes that, "it is not clear that their [Russian] economy will be able to maintain the personnel, infrastructure, and financial resources required to operate these facilities [Wald, 1993]."

The most promising solution to this immediate proliferation threat is to store the fissile plutonium at a single facility with high levels of physical protection with safeguards provided by the International Atomic Energy Agency. The plan presently being discussed is to construct an interim plutonium repository in the Siberian city of Tomsk that will house the excess plutonium coming from dismantled Russian nuclear missiles. However, local opposition to the facility may prevent its construction at this location.

The United States has taken significant steps to help mitigate the proliferation risks in the former Soviet Union by adopting a series of legislative actions, beginning with the *Soviet Nuclear Threat Reduction Act of 1991* (P.L.102-228). In this action, Congress found that:

> [T]he profound changes underway in the Soviet Union pose three types of danger to nuclear safety and stability, as follows: (A) ultimate disposition of nuclear weapons among the Soviet Union, its republics, and any successor entities that is not conducive to weapons safety or to international stability;(B)seizure, theft, sale, or use of nuclear weapons or components; (C) transfers of weapons, weapons components, or weapons know-how outside the territory of the Soviet Union...[I]t is in the national security interests of the United States (A) to facilitate on a priority basis the transportation, storage, safeguarding, and destruction of nuclear and other weapons in the Soviet Union... [PL1, 1991].

Subsequent legislation, including the *Freedom For Russia and Emerging Eurasian Democracies and Open Market Support Act of 1992* and the *National Defense Authorization Act For Fiscal Year 1993*, solidifies U.S. support of Russian dismantlement and storage efforts. Over $800 million has been set aside to "help prevent proliferation of weapons technologies and the dissolution of the technological infrastructure of those states..." [PL1, 1992].

Long-Term Plutonium Disposition

There are a variety of processes which have been suggested to render WGPu less attractive for reuse in weapons. It is now common practice to refer to these process as "disposition options", and we adapt this terminology here.

Reviews of such options have been undertaken by the Congressional Office of Technology Assessment(OTA) and the National Academy of Sciences (NAS). The OTA report was issued in September, 1993, and the NAS study will be released sometime in early 1994. In addition, President Clinton, in his September 27, 1993 nonproliferation policy statement, directed the U.S. government to undertake a comprehensive study of plutonium disposition options. The following disposition options are considered to be the most feasible:

1. Mix the WGPu with radioactive elements such as the high-level waste from reprocessing or non-radioactive elements such as rare earths which are chemically similar to Pu, and incorporate the mixture into a matrix, such as borosilicate glass, to make processing to extract the Pu more difficult. Store the material in either an above-ground

or deep geologic facility, depending on whether the Pu is considered to be waste, or to leave open the possibility of future reuse in reactors.

2. Burn the WGPu in existing nuclear reactors long enough to shift its isotopic composition to that characteristic of RGPu, and in the process, create a significant radioactive barrier to direct use or processing to extract the Pu. This option would allow the plutonium to be used to generate electricity.

3. Burn the WGPu in a nuclear reactor or an accelerator-based neutron source for an extended period so that the plutonium is substantially consumed by fissioning. This process will also generate electrical energy, but would require that some form of plutonium separation and recycling be conducted in order to attain the high burnup required. In the gas reactor, such burnups might be attained on a once-through cycle.

Ultimately, how Russia decides to dispose of the weapons plutonium will be a function of its perceived value as a nuclear fuel, weighed with the incentives afforded by the international community to minimize its inherent proliferation risks. How the U.S. decides to process its plutonium will have an impact on Russia's decision [OTA, 1993], but to what extent is unclear. Unlike Russia, the U.S. has a definite bias against using plutonium as a nuclear fuel in commercial reactors, dating from the Carter Administration, because of the proliferation risk.

1.4 Thesis Objective

This thesis takes a detailed look at one WGPu disposition concept that is being proposed by the Los Alamos National Laboratory (LANL), and considers the concept's technical characteristics in light of U.S. and Russian policy goals. LANL proposes to develop and build a subcritical reactor that uses a high neutron flux derived from an accelerator/target source to totally destroy the WGPu.

The technology assessment consists of three parts. In Part I, (Chapters 1 and 2) the international security problem posed by the WGPu stocks is discussed. Current efforts by the United States, Russia and the international community are examined to determine what goals, or policies, define the boundary of acceptable plutonium disposition strategies. With the overall policy objectives defined, Chapter 2 further distills these goals into three central

technical attributes from which a metric for comparing various technology concepts can be developed.

Then, in Part II (Chapter 3), a framework is developed and used to facilitate the technical discussion of the LANL concept. This framework is designed to allow the analyst to "map" each policy-defined technical requirement to a specific concept characteristic. In this analysis, each major component of the ABC concept is examined in detail to determine its feasibility and technical maturity within the context of a plutonium burning concept.

Finally, using the technical attributes in Part I, Part III (Chapter 4) examines the LANL concept on the merits of its claimed benefits as a plutonium burning reactor. The fundamental question asked during this discussion is: given the goals of plutonium disposition, is the LANL concept a sensible technology to develop? The short-term and long-term implications of developing the LANL concept are examined.

Chapter 2

Technical Requirements for Processing Weapons-Grade Plutonium

The United States has recognized the need to provide safe storage for the Russian excess WGPu until a permanent disposal option can be implemented, and has promised funds to assist Russian efforts to construct an interim storage facility. In addition, long-term disposition issues are being considered, and the process of evaluating alternative disposal processes for both U.S. and Russian plutonium has begun. However, the rationale for plutonium disposition and the best method to accomplish it is viewed differently in the U.S. and Russia. The United States is driven primarily by nuclear proliferation concerns, while Russia emphasizes the importance of using plutonium as a nuclear fuel.

These diverging viewpoints on the value of the WGPu is caused by a fundamental difference in what is viewed as the optimal nuclear fuel cycle. Concerns that a plutonium fuel cycle poses too great a risk of nuclear weapons proliferation, and a lack of clear economic benefit, prompted the United States to abandon its efforts to establish a plutonium fuel cycle in the 1970s [WB, 1992]. However, Russia, along with several other European countries and Japan, has aggressively pursued the use of recycled plutonium in nuclear fuel cycles. In summary, the U.S. believes that the weapons plutonium constitutes a net cost that must be minimized, while Russia believes that plutonium, as fuel, has a net positive value. With a long-standing energy policy that includes a plutonium fuel cycle, it would be politically

difficult for Russia to dispose of its weapons plutonium stock without deriving some real or perhaps *perceived* benefit from it. Though it does not presently have the money to build the necessary plutonium-burning facilities or the experience to operate them, Russia is determined to pursue a plutonium economy [Eco, 1993].

Regardless of these differences, there is a need for reciprocity in the amount of plutonium that is processed in each country, and the extent to which it is irreversibly changed from weapons grade quality. This requirement will likely drive each country to similar disposition methods. As an example of irreversibility, consider a scenario in which the plutonium is simply stored in its pure form. Because it can be taken out of storage and reused in a nuclear device with little or no processing, this constitutes a completely reversible disposition process. In contrast, if the plutonium is transmuted in a reactor for a long period, little plutonium would remain, resulting in a completely irreversible process.

The following discussion attempts to identify what attributes of a plutonium disposition process best satisfy the need to reduce proliferation risks, while meeting the specific requirements of the Russian and United States governments. Although neither country has formulated specific technical requirements for plutonium processing, understanding these key attributes can serve as a guide to discussing what plutonium disposal processes are reasonable.

2.1 Disposition Process Parameters

2.1.1 Plutonium Disposition Time Scale

To understand the time scales and plutonium quantities involved in weapons dismantlement, consider as an example Russian dismantlement and disposition strategy shown in Figure 2-1 (Russian plutonium data was taken from von Hippel *et al.* [vMF$^+$, 1993]). To reduce its strategic nuclear arsenal from 15,000 warheads to approximately 3,500 by the year 2003, in accordance with START agreements, Russia will have to increase its dismantlement rate from the present level of 1000 warheads per year to 1650 [vMF$^+$, 1993]. Assuming that each warhead contains four kg of plutonium, the level of plutonium contained in temporary storage will steadily increase to 100,000 kg by 2003.

However, this level will begin to decrease as plutonium processing begins. The starting date for processing of 2005 is based on a study conducted by Omberg and Walter

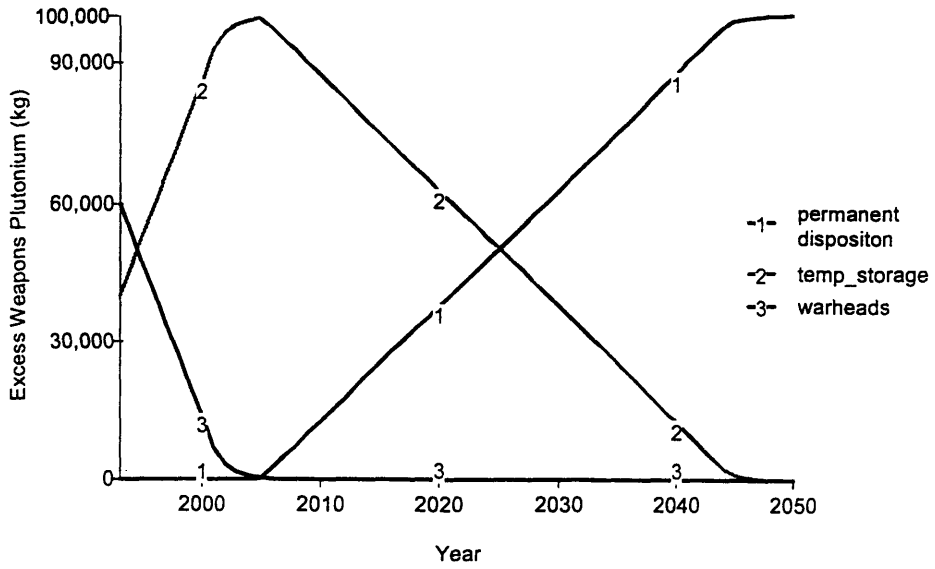

Figure 2-1: Example scenario of plutonium disposition in Russia

Implementation Year	Processing Capacity (MT/year)								
	5.0	4.5	4.0	3.5	3.0	2.5	2.0	1.5	1.0
2005	2027	2029	2031	2035	2039	2045	2056	2073	2105
2010	2032	2034	2036	2040	2044	2050	2061	2078	2110
2015	2037	2039	2041	2045	2049	2055	2066	2083	2115
2020	2042	2044	2046	2050	2054	2060	2071	2088	2120

Table 2.1: WGPu processing "completion year" as a function of capacity and processing start date

[OW, 1993]. In this study, projected disposition start dates ranged from the year 2000 through 2020, depending on the maturity of the technology. For example, processing that involves mixing plutonium with natural uranium (MOX fuel) as a means of burning plutonium in existing light water reactors could be started by in 2005, while new technologies such as the accelerator-based plutonium burner would take in the range of 15 years to develop. Further, assuming that the 100 tons of WGPu will be processed in 40 years, or 2.5 MT/year, the process will be completed by the year 2045.

The time required to process 100 metric tons of plutonium is a function of the process start date and processing capacity. Table 2.1 shows the completion-year for several different combinations of process start dates and process capacity. The assumed processing capacity ranges from 1.0 MT/year to 5.0 MT/year, while the processing start date is varied between years 2005 for mature technologies to 2020, for advanced reactor concepts.

First, note that as the implementation year is delayed, the completion year is merely shifted further in time by the same amount. For example, for a capacity of 1.5 MT/year,

shifting the process start date from 2010 to 2015 results in a shift in completion date by 5 years. Completion time is also a function of processing capacity. For example, given a start date of 2005, a change in capacity from 1.5 MT/year to 2.0 MT/year shortens the total process time by 17 years, from year 2056 to 2073.

The total processing capacity is the product of the unit capacity and the number of units implemented. For example, given a total processing capacity of 2.5 metric tons per year, 7 LWRs using a core containing 1/3 MOX fuel (1200 MWe per reactor) would be needed. However, a Pebble Bed Reactor technology (PBR) would require only three reactors at 630 MWe each [OW, 1993].

The limiting factor for total processing capacity is likely to be an economic one. For the capacity quoted in the Omberg study, the associated capital investment ranged from less than $5 billion to several tens of billions of dollars, depending on the maturity and required level of development necessary to implement the concept [OW, 1993]. Given this, it is likely that, at least for reactor technologies, the total capacity implemented will not exceed 2.5 MT/year. Thus, for the disposition options studied, there is no short term processing strategy. Whether processing begins in 2005 or 2020, a significant amount of the weapons plutonium will remain in storage until approximately 2060. The OTA [OTA, 1993] concludes:

> Significant time will be required for making disposition decisions and formulating policy; for planning, designing, funding, building, and testing even the most available technology; for gaining regulatory and public acceptance; and for actually processing quantities of materials.

2.1.2 Barriers To Prevent Theft by Terrorist Groups

The Department of Energy uses the concept of *attractiveness level* to categorize nuclear materials for control and inventory accounting [DOE, 1988]. The attractiveness of a nuclear material, from a proliferator's view point, is a function of the difficulty of processing and handling required to convert it into a nuclear explosive device. Nuclear facility safeguards, processing, accountability measurements, are designed according to this measure. As an example, assembled weapons and test devices require little or no processing to achieve weapons capability; thus, they are assigned an attractiveness level of "A." Each facility with material at this level is required to have an "extremely high probability of preventing

a single insider from gaining control of a weapon or test device." Likewise, plutonium in the form of pits, major components, buttons, ingots, and directly convertible metals have been assigned an attractiveness level of "B." A facility with material at this level is required to have, "an extremely high probability of detecting abrupt theft of an item containing five or more formula kilograms of SNM [Special Nuclear Materials]."

However, a more direct way to view proliferation resistance is to envision deterrents to the use of weapons materials as barriers between it and the proliferator. There are three types of barriers to consider: institutional, those offered by the processing/storage facility, and those due to the material form of the plutonium. Institutional barriers, in this context, refer to controls offered by IAEA. The United States and Russia have both agreed to IAEA jurisdiction over the excess plutonium stocks in their countries. The second and most obvious barrier is the security offered by the facility where the plutonium is stored or processed. The third barrier is embodied in the nuclear material's material form, which determines the extent of handling and processing required to make it usable in a weapon. Material barriers constitute the last and most important part of a controls strategy and is a function of the processing technology used.

Changing the Physical Properties of Plutonium

The material barrier category can be further divided into four specific material characteristics that make the material more or less attractive as weapon material: isotopic composition, chemical form, radiation level, and quantity. Critical mass and yield of a plutonium fission fission weapon is a function of the isotopic quality of the plutonium and the design of the weapon [WT, 1974].

Figure 2-2 illustrates the various material barriers that are available, depending on what processing technology is employed. For example, one process concept suggests that plutonium can be made as resistant to diversion as commercial spent fuel by mixing the WGPu with high-level waste from reprocessing and placing this composite into glass logs for geologic disposal [vMF$^+$, 1993]. In this scenario, two barriers are employed: radiation and chemical. The main disadvantage to this option is that the isotopic form of the plutonium is unchanged.

Among fission reactor concepts, there are two possible strategies that that can be taken in creating material barriers to reuse of the plutonium in weapons. The first is represented

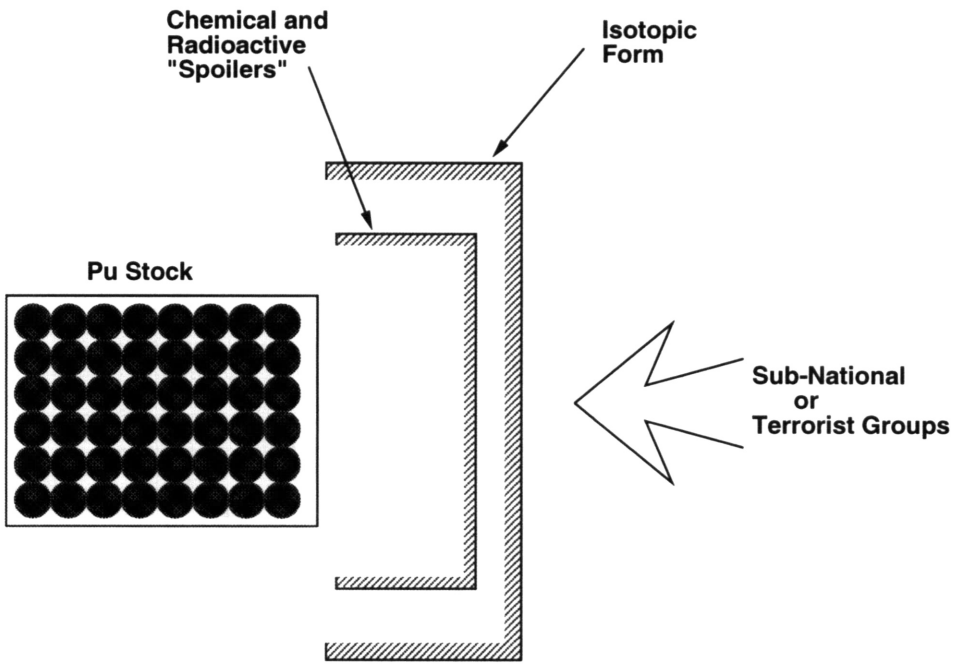

Figure 2-2: Material barriers that hinder attempts to steal Pu for weapons use

by the light water reactor concept. Light water reactor proponents advocate burning the plutonium as a plutonium-oxide/uranium-oxide mixed fuel (MOX). In this case, the Pu^{239} would fission at the same time new plutonium is generated when the U^{238} captures a neutron. At the end of the process, a significant amount of the plutonium remains in the spent fuel. However, a greater percentage of higher plutonium isotopes are present, which reduces the quality of the plutonium to a level comparable to RGPu found in commercial spent fuel. Figure 2-3 (from [Cleave, 1974]) illustrates the production and change in isotopic concentrations of plutonium in enriched uranium (2%) for two types of light-water reactor designs. The curves labeled r=0.2 refer to the Boiling Water Reactor design, while the curves labeled r=0.3 correspond to a Pressurized Water Reactor. Although the total plutonium continually builds as the fuel undergoes fissioning, the isotopic character also changes. Thus, the longer the uranium is fissioned, the lower the quality of plutonium, i.e. a lower concentration of Pu^{239}. In addition to a shift in isotopic character, a significant amount of fission products and higher actinides are generated which offer a high radiation barrier. The argument for this strategy is that reducing the quality of the weapons plutonium to that found in spent fuel is sufficient to hinder its use by terrorist groups. However, the amount of plutonium remaining after this process is complete is significant. Omberg

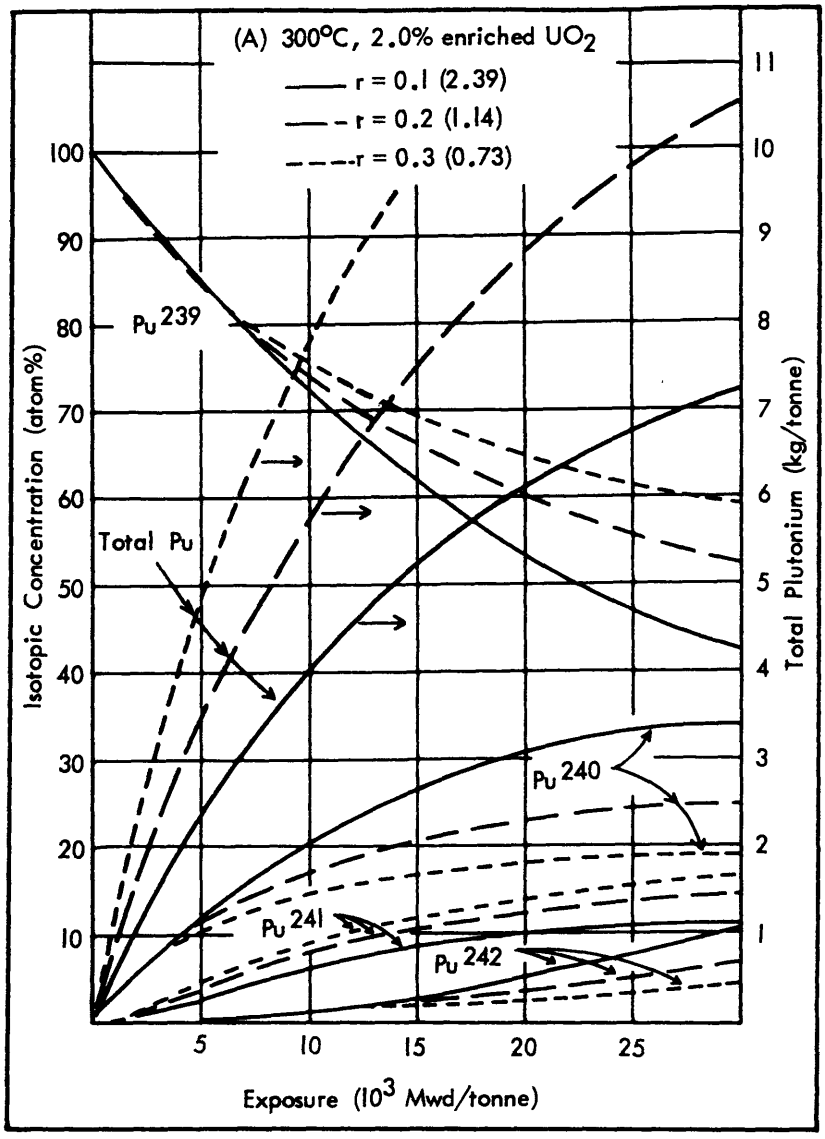

Figure 2-3: Change in plutonium isotopic concentration with fuel residence time

and Walter [OW, 1993] state the percent annihilation of plutonium for these processes is only between 24% and 39%, depending on the specific system.

The alternative view for reactor systems is that the only way to reduce the risk of proliferation is to destroy the plutonium completely, or at least substantially. To accomplish this, reactor systems would have to be designed that would allow the Pu to be burned (fissioned) for a much longer period than is possible using present light water reactor technology. The buildup of actinides and fission product poisons in solid fuel elements prevent high burnup of the plutonium. In some reactor concepts, e.g., the high temperature gas reactor (HTGR), significant burnup is possible on a once-through basis. The more advanced reactor concepts propose to employ a continuous recycling process. For example, a continuous-flow variation of present-day reprocessing is offer by LANL's accelerator-based (ABC) reactor system. This concept would employ a slurry-based fuel that would allow the poisons to be removed on a continuous basis. Omberg and Walter cite the percent annihilation possible for this and other advanced reactor designs in the range from 56% to 99% of the original plutonium [OW, 1993].

The extent to which a disposition technology, reactor or non-reactor, offers one or several of the physical deterrents noted above will determine how resistant the plutonium is to illegal diversion and use. The decision of whether converting the plutonium to a spent-fuel-like form or complete annihilation will be employed must be mandated by policy.

2.1.3 Plutonium as a Fuel

However, the attractiveness of various plutonium disposition options is driven by more than their ability to impart barriers to diversion. Another major factor to consider, particularly in the case of the Russian government, is how well a disposition process is able to extract the plutonium's value as a nuclear fuel [Jaf, 1993]. Based on Russia's commitment to develop a closed nuclear fuel cycle, which it considers as a necessary part of maintaining a credible nuclear capacity [MSR, 1993], the Russian Ministry of Atomic Energy (MINATOM) considers any proposal that treats the weapons plutonium as a form of waste as "heresy" [vH, 1991, Eco, 1993].

There is little doubt that MINATOM is convinced that a closed nuclear fuel cycle is the best means of assuring a stable energy capacity [NE1, 1991, NE2, 1992]. In their view, nuclear power will eventually serve as the primary source of the world's energy, and they desire

to play a significant role in this market as an exporter of nuclear technology, as well as using nuclear power to supply a significant fraction of Russia's energy needs [Nathanson, 1979]. Therefore, a technology like the breeder reactor is an attractive alternative because it uses uranium resources much more efficiently than current reactors by breeding new fuel (plutonium) during operation. Wright [Wright, 1977] calculates that with an installed breeder reactor nuclear capacity base, it is possible to obtain desired electrical capacity levels with 50 times less uranium than with using conventional light water reactors.

However, the United States, not convinced of the benefit of a closed cycle, and concerned that a plutonium-based nuclear capacity would enhance nuclear proliferation, abandoned its pursuit of a closed nuclear cycle. Under the Carter Administration, the US deferred its recycling efforts with the hope of leading the world away from a plutonium-based energy strategy [WB, 1992], however, this policy has only been marginally successful.

Incentives for a Plutonium Economy

Before this shift away from a closed fuel cycle, the concept of using plutonium as a nuclear fuel was a key part of the energy strategy in many countries, including the United States [Suzuki, 1991]. Special fast breeder reactors would be employed to produce more plutonium than is consumed, as well as energy. This process, in the view of many, was the only way to maintain a sustainable nuclear fuel supply in light of a growing nuclear power demand and limited natural uranium deposits [WB, 1992].

This philosophy changed drastically in the 1970s for two reasons. First, the United States and others recognized that the plutonium produced in the light-water reactors, as well as the breeder reactors, was weapons usable. Second, because the demand for nuclear capacity did not grow as expected, and therefore, the uranium stores would be sufficient to feed expected capacity, the unit price of uranium fuel dropped substantially. As a result of this price decrease, coupled with the higher-than-expected processing costs for plutonium, the economic attractiveness of a plutonium-based fuel cycle quickly diminished [FTL, 1991, BDF$^+$, 1992, Suzuki, 1991].

Figure 2-4 and 2-5 illustrate the once-through and the recycle nuclear fuel cycles, respectively. The unit cost of a fuel element is a function of the material and processing costs. In economic terms, the plutonium recycle is unattractive because of the lower cost of natural uranium coupled with the high fabrication costs associated with plutonium bearing MOX

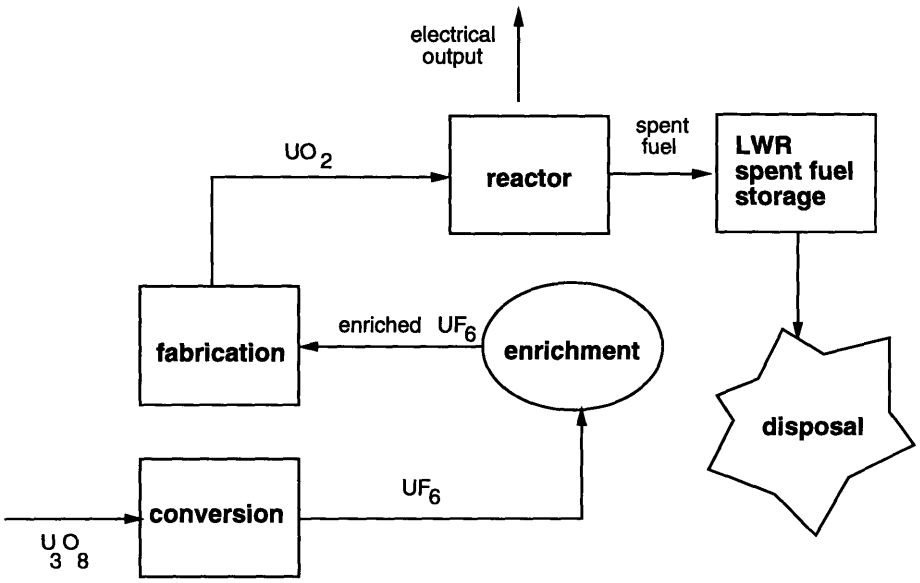

Figure 2-4: Once-Through Nuclear Fuel Cycle

Transaction	*Unit Cost* ($/kg)	*Mass Flow* (kg/kg)	*Direct Cost* ($/kg)	*Carrying Charge* ($/kg)
ore purchase	65	5.96	387	271
enrichment	130	4.8	624	312
fabrication	150	1.01	151.5	61
storage	100	1.0	100	-30
disposal	250	1.0	250	-75
subtotal cost			1513	539
total cost				2052

Table 2.2: Plutonium Fuel Recycle with MOX fuel

fuel. Lester [Les, 1992] gives a sample calculation to demonstrate this point. Tables 2.2 and 2.3 list the component costs and overall unit cost of an all-uranium fuel and MOX fuel. In this example, the uranium fuel is 1,858 $/kg less than the MOX fuel, for an equivalent capacity fuel benefit. Employing a MOX fuel would increase the unit cost of fuel for a LWR by nearly 90%.

Different Goals in Processing Plutonium

Without the need to rationalize use of plutonium as a fuel, the United States remains focused on looking for process technologies that will *effectively* dispose of WGPu stocks in the shortest time possible and at a minimum cost. Because of present uranium fuel prices, the electrical energy generated by processing plutonium in reactor-based systems, as defined

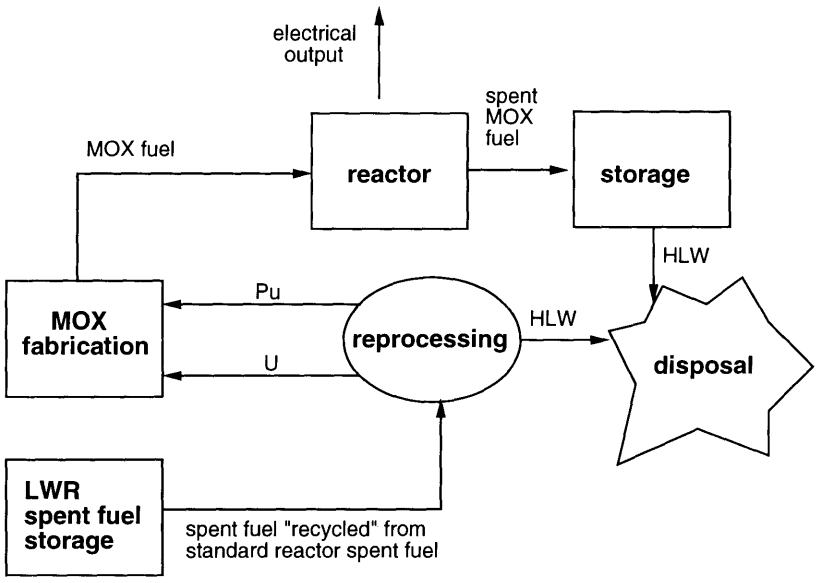

Figure 2-5: Plutonium Fuel Recycle

Transaction	*Unit Cost* ($/kg)	*Mass Flow* (kg/kg)	*Direct Cost* ($/kg)	*Carrying Charge* ($/kg)
credit: reduce storage	350	3.33	-1155	-578
reprocess	750	3.33	2498	1249
disposal	250	3.33	833	250
MOX fabrication	550	1.01	556	222
uranium credit	65	31	-150	-60
storage	100	1	100	-30
spent fuel disposal	250	1	250	-75
subtotal cost			2932	978
total cost				3910

Table 2.3: Fuel cost comparison assuming 1 kg reload fuel

by the MOX fuel cycle, would serve only to offset the costs of the disposition process. Thus, in economic terms, plutonium as a nuclear fuel has a negative value.

Regardless of the economic disincentive to use an MOX fuel, Russia is still determined to use its weapons plutonium to power reactors, which limits the options it will consider.

Foreign interests more sympathetic to Russia's desire for a plutonium economy have also been involved in the disposition issue. Siemens, a German firm, proposes to build a MOX plant in Russia that could fabricate about 6-7 tons of reactor grade plutonium per year [Dahlburg, 1992]. Japan has proposed to construct a specially designed version of its plutonium breeder reactor that consume up to 2 tons of plutonium per year [Sov, 1992].

The cost/benefit balance of plutonium disposition

To understand the trade-off between extracting the potential energy value of the WGPu and the cost of processing it for permanent disposal, it would be helpful to consider a general cost/benefit curve as shown in Figure 2-6. In this graph, the cost per kg of plutonium that undergoes processing is shown on the x-axis. The y-axis is a measure of the unit value of energy produced by the process, and is also in dollars per kg processed.

The 45^o line constitutes the break-even point between these two factors, and indicates where the unit cost of disposition processing is exactly equal to the benefit derived from its use as a fuel. Assume, for example, that the unit cost of disposition is $ 1000 per kg for a particular technology. The only way this scenario would "pay for itself" is if the total value of the energy produced were equal to or greater than the cost to produce it. However, since the market unit price of electrical energy reflects the lower cost of uranium fuel, the derived benefit from burning the more expensive plutonium-based fuel will never exceed the break-even point. This, of course, assumes that the price of uranium fuel remains low. Thus, the region above the break-even line indicates a net profit, and the region below it indicates a net cost of production.

As an example, consider two disposition process technologies indicated by points A and B. Assume that both technologies result in the same level of proliferation barriers after processing. Although the unit cost of processing for technology A is higher than B, since technology B does not fission the plutonium and therefore extracts no electrical credit, the net cost of processing it is exactly the same as for A. The only difference in this example is that one technology produces electrical energy and one does not. On this point, von Hippel

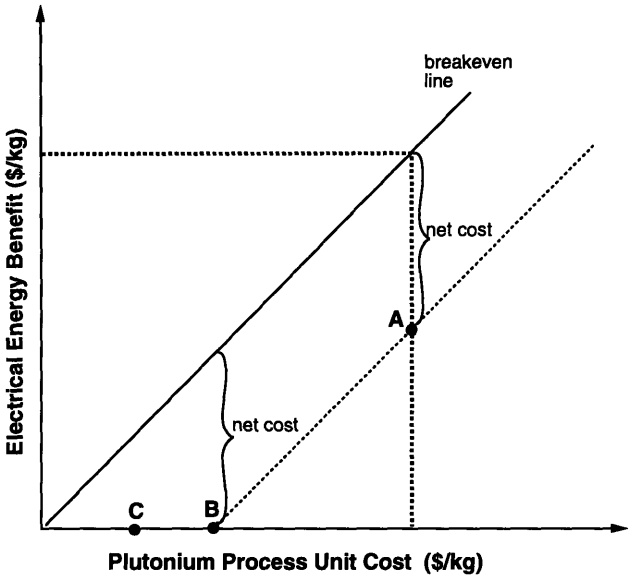

Figure 2-6: Plutonium Disposition Cost-Benefit Plot

et al. [vMF$^+$, 1993] write:

> Although disposal of plutonium with radioactive waste would forego the electricity it could generate, this loss is insignificant in the larger context. At present uranium and plutonium prices, plutonium will not be an economic fuel for at least several decades.

It is clear that given the choice between A and B, Russia would choose A because it uses the plutonium as a fuel for powering reactors. In fact, it seems that Russia is not willing to consider any technology that does not extract some economic value, even if the net unit cost of processing is greater for non-energy processes. A case in point is the option to mix the plutonium with radioactive waste, which is believed to be less costly, or comparable to, than the reactor alternatives [OTA, 1993]. This option can be represented as point C in Figure 2-6. Russia's outright rejection of such proposals indicates that their primary concern is not to reduce the risk of proliferation but to promote the longer-term goal of developing a plutonium fuel cycle. Christopher Paine, a senior researcher with the Natural Resources Defense Council, expresses concern over this point:

> The Russians are very enthusiastic about a plutonium fuel cycle that involves reprocessing the fuel many times. But we would not want to subsidize a plutonium fuel cycle under the guise of disarmament. If you legitimate plutonium as

a civilian fuel, your are legitimizing the possession of plutonium fuel facilities by any country that wants them and thus the possession of plutonium that could be used for nuclear weapons" [NS, 1992]

2.2 Designing a Strategy to Meet the Adversary

From a non-proliferation standpoint, the primary function of a disposition process is to transform the weapons plutonium into a form that will prevent terrorist groups and other subnational groups from obtaining and using it for nuclear weapons. There is much less concern that Russia would extract the plutonium from permanent storage simply because they already have the technical and industrial means of producing any amount of plutonium desired, and with a large, active nuclear arsenal, the excess stored plutonium makes little difference.

The primary challenge lies in preventing diversion attempts by terrorist groups who could not otherwise produce fissile materials. For example, Giles [Giles, 1983] reports one attack that was launched by political extremists in the once-Soviet republic of Azerbaijan on a nuclear warhead facility near Baku. Fortunately, their efforts were thwarted by Soviet troops. It is clear, then, that the risk of proliferation will remain high as long as the fissile material remains in its weapons form. Frank von Hippel *et al.* [vMF$^+$, 1993] write that "so long as the recovered materials remain in forms easily converted back to weapons, their existence will erode confidence in the disarmament process and raise dangers of diversion to non-nuclear nations and terrorists."

Given that our goal is to prevent the terrorist or sub-national group from gaining access to and using the plutonium, what processing characteristics should be considered when evaluating alternative plutonium disposition strategies? First, disposition processing must impart an adequate level of physical barriers to prevent terrorist groups from using the material if some amount were acquired. The baseline comparison for this requirement is commercial reactor spent fuel, which is stored in large quantities throughout the world. The second requirement is that the technology must satisfy, at least to some degree, Russia's desire to extract economic benefit from the disposition process. Alternatively, the United States is most concerned with national security issues, and would not desire to encourage a plutonium economy.

Chapter 3

The Accelerator-Based Plutonium Transmutation Concept

3.1 Introduction

It is critical that specific technology development goals be defined by, or reconciled with, established governing policies prior to evaluating alternative concepts. This is especially true for very large and complex technology development efforts, such as a plutonium processing and disposal facility, where the required investment is likely to run into several billions of dollars. In Chapter 2, it was shown that the most important attributes to consider for a plutonium processing technology include: (1) the degree of proliferation resistance, i.e., the barriers against possible reuse of the Pu in weapons, (2) the implementation time and capacity of the technology, and (3), to a lesser extent, the amount of economic benefit derived from the process. In this chapter, we use these defined attributes as a framework in which to discuss and evaluate a plutonium processing concept that has been proposed by the Los Alamos National Laboratory. Their innovative concept is a significant departure from the present reactor technology base, and promises an operating performance that goes well beyond other concepts defined for WGPu disposal. The following discussion provides a bridge between this plutonium processing concept and the goals defined by US and Russian policy in order to determine whether research and development support should be given to this proposal.

The basic concept behind the Los Alamos proposal is to burn the weapons-plutonium

in a reactor-like subcritical system that relies on a linear accelerator to produce the large exogenous neutron source required for subcritical operation. The possible benefits of using a subcritical fissioning system may include: increased safety due to the subcritical nature of operation, continuous processing of fission byproducts, and a high plutonium/actinide burn rate per volume [ADL, 1992].

Accelerator-based systems have long been considered for applications where neutron production is of primary concern. Beyond particle-physics and materials research, these systems have been proposed as a means of breeding fissile Pu^{239} and U^{233}, transmuting actinide and fission product waste; and as advanced fission reactors [SPT+, 1978, Schriber, 1978, Bowman+, 1992, Newman, 1991]. Although the interest in using accelerators for these purposes is long-standing, the required investment in research and development to build these systems has, thus far, been greater than the perceived benefit of their use. Thus, progress has been slow, and these technology concepts remain relatively undeveloped.

Of particular interest in this discussion is concept of the transmuting nuclear waste, such as actinides, e.g., americium and curium, and long-lived fission products such as technetium. Both reactor-based and accelerator-based concepts have been considered as a means of transmuting radioactive waste streams from defense and commercial nuclear processes. However, resistance from some in the nuclear community to what is perceived as an unwelcome distraction from geologic disposal efforts, as well as proliferation analysts concerned about the ability of such processes to separate Pu, has prevented significant development work on these concepts from taking place. Even if partitioning of the waste and subsequent transmutations were developed to augment the US geologic disposal strategy, skeptics argue that the benefit of transmuting the long-lived constituents of these wastes before disposal in a repository does not warrant their associated development costs. In a recent report evaluating the availability of various transmutation technologies, Ramspott *et al.* [RCHP, 1992] conclude that:

> There remains no cost or safety incentives to introduce P-T [partitioning and transmutation] into the HLW management system. The absolute risk from radioactivity from a repository is very low, and the actinides that P-T would remove or reduce do not contribute significantly to that risk....

It is within this clearly unfavorable environment toward waste transmutation, that Los Alamos National Laboratory proposes their plutonium-burning concept, which is based al-

most entirely on previous development work with their Actinide Transmutation of Waste (ATW) system. In fact, they highlight the capability of their system to transmute the actinides americium, curium and some long-lived fission products in addition to the weapons-plutonium. It is important, therefore, that the merits of the accelerator-based technology be considered relative to the problem of weapons-plutonium processing and not nuclear waste disposal. Specifically, does the added function of continuously separating and recycling the plutonium and other actinides for near complete annihilation warrant its development costs?

Three questions will be addressed in this chapter:

1. What features of the technology are responsive to the plutonium disposition policy objectives?

2. How does the technology function to accomplish these objectives?

3. What technical issues and concerns affect the probability that the technology can be successfully developed to the capacity required for plutonium disposition?

3.2 Policy Driven Technology Development

In light of these questions, a technology must be evaluated in a manner that shows its connection to policy-driven technical requirements. However, correlating these two worlds is often difficult for complex systems, and often times, technology performance attributes not directly linked to a policy's direction are used as figures-of-merit. In his book, *The Principles of Design* [Suh, 1990] Nam Suh writes concerning this difficulty:

> Clearly, one of the major problems in the political domain is that there are many factions or groups pulling society in many different directions, therefore making it difficult to *design* tasks, organizations, and approaches. It takes a skilled politician to prioritized goals and engineer an agenda despite the obvious conflicts.

Thus, an effective technical evaluation must have two key elements. First, the evaluation must be framed around a clear definition of policy driven technical requirements. Second, the technology concept must be clearly defined relative to these requirements.

Figure 3-1 illustrates a hierarchical design framework that can be used to accomplish this [Suh, 1990]. First, at the highest level of a design evaluation, the "policy directive" is

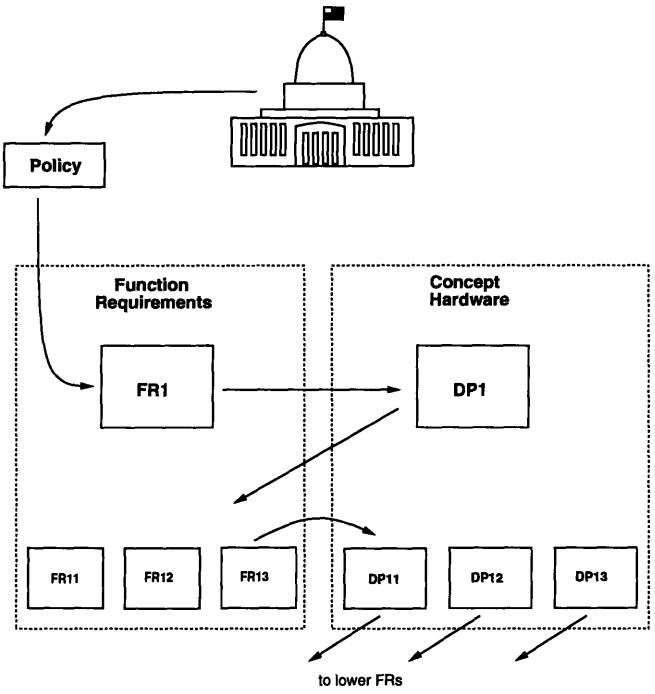

Figure 3-1: Design Requirements derived from policy

translated into a statement of objective, also called a *functional requirement* (FR), which constitutes the overall objective of that technology development effort. Correspondingly, the *design parameter* (block labeled DP1) describes, in general terms, the technology concept (or hardware) that will be considered as a solution to the highest level functional requirement.

At the next level, the minimum number of functional requirements that describe the concept are defined (FR11, FR12, and FR13). For a designer these requirements would serve to guide decisions between alternative hardware subcomponents (DP11, DP12, and DP13). However, for concepts that have already been defined, this step establishes the "physics" of the system and allows an evaluator see the link between the overall policy goals and how the concept functions to meet them. The process of defining the concept into further levels of detail is continued until the system is sufficiently characterized to

evaluate its merits as a solution to the main functional requirement, and, therefore, the policy objective.

The LANL accelerator-based transmutation system can be treated in this manner. First, polices with regard to plutonium disposition must be restated in terms of a functional requirement. As discussed in chapters 1 and 2, the United States has pursued a policy that includes funding efforts to find safe storage capacity for Russia's WGPu. In the longer term, however, the plutonium will have to be processed into a form that makes it more resistant to use in a nuclear device. Thus, this policy goal can be restated as a functional requirement:

> FR1 $\equiv$ The plutonium must be processed such that physical barriers against its use in a nuclear device are maximized to the greatest extent reasonable. The physical barriers may include one or all of the following: Pu^{240} or higher plutonium isotopes in high concentrations (isotopic barrier), radioactive fission products and/or chemical poisons, substantial annihilation (extreme case of isotopic barrier)

At this highest level, the choices in technologies vary between fissioning and non-fissioning processes. However, since the accelerator based system is a reactor technology, the systems hardware can be described in the following way:

> DP1 $\equiv$ A fission reactor that transforms Pu^{239}, through a fission and neutron capture process, into other isotopic species, actinides, and fission products to reduce its usefulness as a fissile material in nuclear weapons.

Now, as the framework proceeds to further levels of detail, any reactor concept could be brought into the discussion and presented. However, this analysis will concentrate on the LANL proposal. In the following section, the Accelerator-Based Converter (ABC) concept will be discussed, briefly, to provide the necessary background for the more detailed analysis that is to follow.

3.3 Accelerator-Based Plutonium Burner Concept Description

3.3.1 System Overview

The basic principle behind the LANL ABC is to fission WGPu by circulating it, in an oxide slurry form through a high thermal neutron flux while continuously removing fission

byproducts that would normally reduce the neutron economy of a fuel system. The oxide slurry is circulated through one or more heat exchanger to remove the fission energy, which is then converted to electrical power for on-site and off-site distribution.

There are two fundamental differences between the ABC concept and existing reactor designs. First, existing reactor technology use fixed solid fuel elements which are cooled by light or heavy water which is circulated through the core. The ABC concept, on the other hand, uses the oxide slurry fuel system which is circulated in and out of the "core region" for cooling. Second, unlike all present day power reactors, the ABC system operates in a subcritical state, that is, the neutrons which cause fissioning are not self sustaining and must be replenished from sources outside the core. Standard reactors operate in a steady state in which the number of neutrons created by fissioning, at some time, is exactly equal to the number of neutrons absorbed by materials in the reactor.

The ABC concept, shown in Figure 3-2 [ADL, 1992], consists of three distinct subsystems. First, in the accelerator/spallation subsystem, high-energy protons which are generated by a medium energy linear accelerator impinge on a heavy metal target which, in turn, generates numerous (approximately 20) neutrons per proton-metal collision, also known as a spallation event. In the ABC baseline concept, a medium energy, 100% duty cycle, linear accelerator (linac) creates and accelerates a 190mA proton beam to an energy level of approximately 800MeV prior to impacting the heavy metal target. The power density on the target is approximately 1.9 MW/L.

The principal function of the spallation target is to convert the high intensity and high-energy proton beam into neutrons and deliver them to the blanket slurry fuel. When a proton collides, or spallates, with a heavy target atom, several nucleons, including protons, are emitted into the surrounding slurry blanket. The ABC baseline system uses tungsten as the primary spallation material and lead as the secondary heavy metal. Tungsten is used primarily because of its high density and ability to withstand high energy densities without structural failure. Lead is used to "catch" the protons that are not absorbed by the tungsten material. Lead is effective for this purpose because unlike tungsten, it does not absorb neutrons, thus, more neutrons are able to enter the slurry. The low melting point for lead restricts its use as the primary spallation material.

Second, the heart of the accelerator-based transmutation concept lies in the actinide slurry fuel subsystem which serves as the fuel and transport mechanism for the plutonium

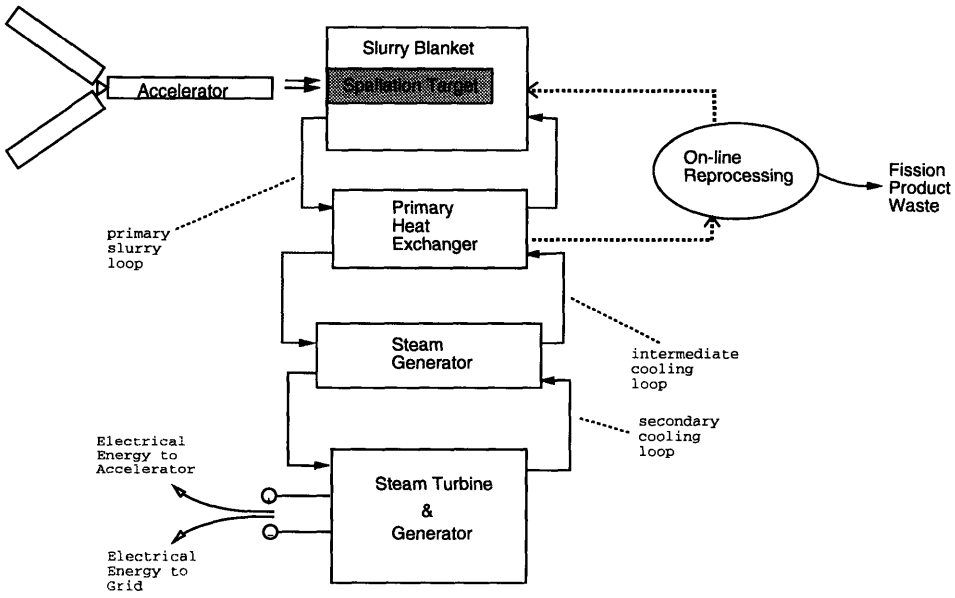

Figure 3-2: Accelerator-Based Subcritical Plutonium Transmutation Concept

and other actinides, and includes the primary actinide slurry loop and a slip stream reprocessing loop. In simplest terms, the ABC slurry can be defined as an aqueous suspension of actinide oxides, including Pu, Np, Am, and Cm and various fission products. Initially, the slurry contains only PuO_2 and heavy water, D_2O. However, in a high neutron flux environment, fission products and higher actinides begin to build-up in the system.

To maintain a sufficient proportion of fissile material in the slurry, a small slip stream (approximately 7% of the total flow) is extracted from the slurry loop and chemically processed to remove unwanted actinides and fission products.

The third segment of the ABC transmutation system is a standard reactor cooling system. Since the "fuel" of the ABC is fluid, fission energy is extracted from the slurry loop through two counter-flow, shell-tube heat exchanger, with the slurry flow on the tube side.

The fission energy is transferred to a heavy water secondary system similar to a concept used in the CANDU reactor concept [CISK, 1992]. The D_2O slurry flows into the slurry heat exchanger where the thermal energy is transferred to an intermediate cooling loop. Finally, the thermal energy is used in the steam cycle to drive a standard steam turbine

before being returned to the steam generator [Nero, 1979]. The baseline ABC configuration specifies a 2140 MW_{th} capacity output, with a 28% thermal efficiency.

Subcritical Nuclear Systems

Thus far, the discussion has alluded to the requirement that the accelerator is needed to transmute plutonium and higher actinides in a sub-critical system. To better understand this concept, a brief discussion of "subcritical" neutron multiplication is now presented.

In a generic chain-reacting system, the steady-state neutron population is the key determinant of the rate at which the fissile material undergoes fissioning, and the rate at which neutrons are absorbed by fertile materials. Since the average lifetime of a neutron is extremely short (10^{-6}s), a steady-state population implies their continual creation and destruction. The time between when a neutron is released in a fission event to the time of its absorption by another atom, or loss outside the system, is considered one neutron life-cycle.

There are several possible interactions that a neutron can undergo in its lifetime. The neutron can be absorbed by an atom causing the atom to fission. In a fission event, the nucleus splits into two or more smaller fragments (called fission fragments or fission products) while releasing several neutrons and energy. In some cases, however, the neutron is absorbed by an atom but does not result in a fission. This reaction is called a neutron capture. Finally, a neutron can simply leak out of the system. To what extent these processes occur in a particular reactor system is dependent on the size and shape of the system, the types and amounts of material present, and the energy of the neutron.

A common way to describe the state of the neutron population in a chain-reacting system is to derive its effective neutron multiplication factor, denoted as K_{eff}. In general, K_{eff} can be described as the fractional change in neutron population per generation; thus, if $K_{eff} > 1$, the neutron population is increasing (supercritical condition), or, if $K_{eff} = 1$, the population remains constant(critical condition), but, if $K_{eff} < 1$, the neutron population is decreasing (subcritical condition). For example, a K_{eff} of 1.05, implies that a system will be increasing by 5% every neutron lifetime [Hughes, 1953].

Specific system characteristics determine the K_{eff}. The *Six Factor Formula*, as it is commonly called, defines K_{eff} as a function six dominant reactor parameters that either causes an increase or decrease in neutron population [DE, 1990, Hughes, 1953] and can be

written as:

$$K_{eff} = \eta \varepsilon p f L_f L_{th} \tag{3.1}$$

The *Reproduction Factor*, η, represents the number of fission neutrons that result from an absorption reaction by a fuel atom. This factor considers both probability of capture to fissioning and the neutron yield from a fission event. The *Fast Fission Factor*, ε, is the increase in fission neutrons resulting from fast neutron fission. Typical values range from 1.01 to 1.03. The *Resonance Escape Factor*, p, describes the probability that neutrons will *not* undergo resonance capture reactions (a non-productive absorption) as they slow down to thermal energy level. The *Thermal Utilization Factor*, f, describes the probability that the neutrons that reach thermal energy will be absorbed by fuel atoms. Not all of the absorption reactions in the system are with fuel atoms, thus this factor is always less than one. Finally, the *Fast and Thermal Non-Leakage Factors*, L_f and L_{th} respectively, equate the probability that the neutrons will leak out of the core while slowing down to thermal energy and while thermalized. In both instances, their value is less than one.

There are two conditions where the neutron population in a system is constant. The first, of course, is when K_{eff} is unity. The second case is when K_{eff} is less than one, but with the neutron losses, as described above, replaced by an exogenous neutron source. To describe this mathematically, consider a form of the one-group, or single-energy, diffusion equation as presented by Arthur and Bowman [Arthur, 1992, Bowman+, 1992]:

$$\frac{dn}{dt} = S + \nu R_f - (1+\alpha) R_f - \nu R_f L \tag{3.2}$$

where S is the external neutron source; νR_f defines the neutrons produced per second due to fissioning; $(1+\alpha)R_f$ defines the neutrons per second needed to sustain fissioning; and $\nu R_f L$ is the fraction of total neutrons generated by fissioning that are lost by leakage and parasitic capture.

Rewriting the system reaction rate, R_f, in terms of of neutron population, n, and the *unit* reaction rate, r_f, Equation 3.2 becomes:

$$\frac{dn}{dt} = S + \nu r_f n - (1+\alpha) r_f n - \nu r_f n L. \tag{3.3}$$

Now, consider the specific case where the reactor system is in equilibrium ($\frac{dn}{dt} = 0$) and

subcritical ($K_{eff} < 1$). Solving for the steady-state neutron population, n, Equation 3.3 yields:

$$n = \frac{S}{(1+\alpha)r_f + \nu r_f L - \nu r_f} \tag{3.4}$$

or,

$$n = \frac{S}{r_f[(1+\alpha) + \nu L - \nu]}. \tag{3.5}$$

To understand how the neutron multiplication factor, K_{eff} relates to Equation 3.5, first rewrite K_{eff}, which is defined as the fractional change in neutron population per neutron generation, in terms of neutron production and destruction:

$$K_{eff} = \frac{production}{destruction} = \frac{\nu R_f}{(1+\alpha)R_f + \nu R_f L} = \frac{\nu}{(1+\alpha) + \nu L} \tag{3.6}$$

thus,

$$[1 - K_{eff}] = [1 - \frac{\nu}{(1+\alpha) + \nu L}] = [\frac{(1+\alpha) + \nu L - \nu}{(1+\alpha) + \nu L}] \tag{3.7}$$

Using this result, we see that Equation 3.5 can be rewritten as:

$$n = \frac{S}{r_f[1 - K_{eff}][(1+\alpha) + \nu L]}. \tag{3.8}$$

Finally, noting that the system is in steady-state, and, therefore, the *net neutron loss rate* = *net neutron production rate*, Equation 3.8 can be expressed as:

$$\underbrace{r_f n[\overbrace{(1+\alpha)}^{absorption} + \overbrace{\nu L}^{leakage}]}_{Loss} = \underbrace{\frac{S}{[1 - K_{eff}]}}_{Production}. \tag{3.9}$$

Transmutation Rate

In the previous section, a description of how the neutron population changes as a function system characteristics was given. Now, to calculate the "transmuting rate" of the fissile and fertile materials within the system, consider a condition where the neutron population has reached equilibrium. The rate at which a material undergoes transmutation is a function of the system flux, ϕ, and the cross section for transmutation, σ.

For a given system, the neutron density, $n[\frac{n}{cm^3}]$, is defined as the ratio of the average neutron population and system volume. The neutron flux of the system is simply the

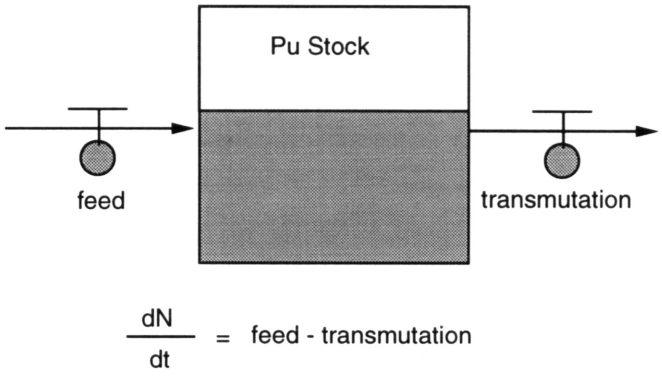

Figure 3-3: The *Feed* and *Transmutation* flow rates determine the level of plutonium at a given instant.

product of neutron density, n, and velocity, v, and represents a measure of particle motion per unit volume [Harms, 1987]. Symbolically, this is expressed as follows:

$$\phi = nv \tag{3.10}$$

Figure 3-3 graphically represents this process for a given stock of plutonium. In this simple system, the rate at which plutonium atoms are fissioned the thermal neutrons (transmutation rate) and the rate at which new plutonium is fed into the system by an external source or created by neutron capture of uranium, determines the level of plutonium at a given time.

We are particularly interested in the transmutation rate portion of this equation because it describes the capacity of a system to process plutonium inventories. The transmutation equation for this system can be written as:

$$TR = \phi\sigma_f N^* + \phi\sigma_c N^* + \lambda_N N^* \tag{3.11}$$

Where the transmutation rate, TR (gm/s) is defined as the rate at which a material undergoes neutron interaction either by (1) neutron absorption (resulting in fission event or simply a neutron capture) by the target atom, or (2), natural decay. The material inventory denoted by N* represents the total number of atoms present in the system. This quantity is a product of the product of atom density, N ($atoms/cm^3$), and system volume, V (cm^3). Fission cross-section, $\sigma_f(cm^2)$, is a measure of the probability that an atom will absorb a neutron and result in a fission event. The capture cross-section, $\sigma_c(cm^2)$, is a measure of

the probability that a target atom upon absorbing a neutron will not fission. As described above, the neutron flux,$\phi, (n/cm^2 s)$, is defined as the average number of neutrons striking a unit area in one units time. The decay constant, λ_N, is defined as the probability per unit time that an atom will decay, and can be expressed by $ln(2)/t_{1/2}$ where $t_{1/2}$ is the time required for one half of the initial atom inventory to decay. Finally, the Feed Rate, $F_N(m^3/s)$, consists of all material flowing into the inventory control volume. This includes sources from outside the reactor system and from neutron interactions of other material species within the reactor system.

The thermal power capacity of the system due to fissioning can be calculated by multiplying the rate of fissioning, $R_f = \phi \sigma_f N^*$, by the energy generated per fission,

$$P_{th} = R_f E_f \tag{3.12}$$

In preliminary calculations, it is recommended that the value to use for energy released per fission is 200 MeV [Lamarsh, 1975].

To illustrate how a fissioning system behaves for a given constant flux level, consider the following: A system containing an initial 10 MT Pu^{239} (only Pu^{239} is considered for simplicity) undergoes fissioning at a flux level of $1x10^{14} \frac{n}{cm^3 s}$ and $1x10^{15} \frac{n}{cm^3 s}$. Further, assume that the feed rate is zero, which implies that the external feed of Pu^{239} is zero and that no uranium is present. Given that the thermal capture and fission cross sections for Pu^{239} are 269 barns $(2.69x10^{-22} cm^2)$and 742 barns $(7.42x10^{-22} cm^2)$ [Lamarsh, 1975], respectively, the plutonium inventory is transmuted as shown in Figure 3-4(a). Figure 3-4(b) shows the associated power curve. The point of interest is how the time required to transmute the initial inventory varies with flux level. At a flux level of $1x10^{14} \frac{n}{cm^3 s}$, it took approximately 1.3 years to consume the stock of Pu^{239}, whereas at a flux level of $1x10^{15} \frac{n}{cm^3 s}$, this process took only a fraction of a year.

Actinide and Fission Product Buildup

For the example given above, it was assumed that only Pu^{239} was present in the system. In a real reactor system, however, the "fuel" really consists of many isotopes, all of which have particular fission and neutron capture properties. Of particular interest are materials created by the fission process itself.

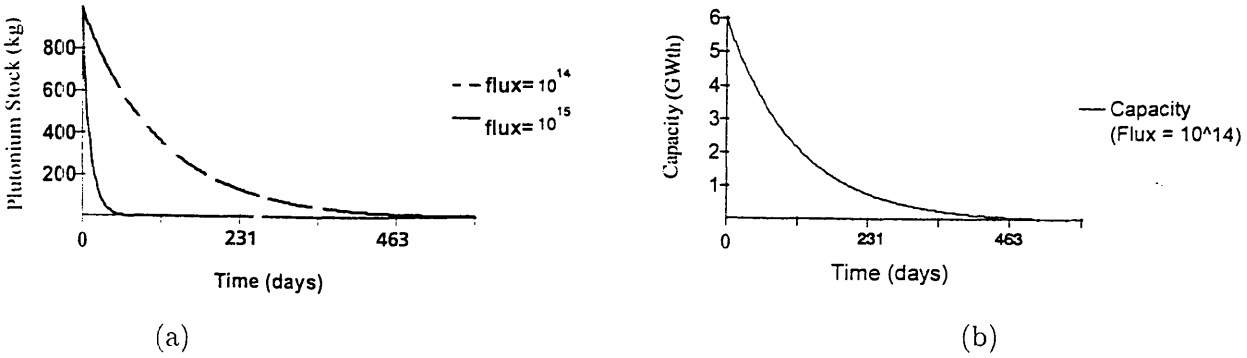

Figure 3-4: Plot of Pu^{239} Level(a) and Corresponding Power Level (b) given a fixed neutron flux of $5x10^{14}\frac{n}{cm^3 s}$.

The products of a neutron absorption are of two general types. If an original fuel atom fissions, the nucleus spits into two smaller particles called fission products. Some fission products are strong neutron absorbers, and thus reducing the overall neutron population if present in the system. If the neutron is simply absorbed by the fuel atom, the atom is converted to a higher isotope of the same element, and may then beta decay to form a higher actinide. Figure 3-5 shows a partial decay scheme for the higher actinides beginning with plutonium [BPL, 1981]. Fission products are not shown.

Thus, over time, the higher actinides, including Np, Am and Cm, in addition to the fission products, accumulate in the fuel system. As these materials accumulate in the fuel, the *thermal utilization factor*, f, of the system is reduced, which, in turn, reduces the system's K_{eff}. To maintain power for a system that operates below criticality, either the source neutron rate would have to be increased to meet the new demand by parasitic capture, or the neutron absorbing materials would have to be continually removed from the system.

Calculating the Required Neutron Source for the ABC System

To determine the neutron source, S, for the ABC spallation process, the target yield (number of neutrons generated per proton), must first be determined. In general, the yield from a spallation target is a function of the target material and the energy of the incident proton. Fraser *et al.* [FB, 1983] have published target neutron yield data for various materials. For

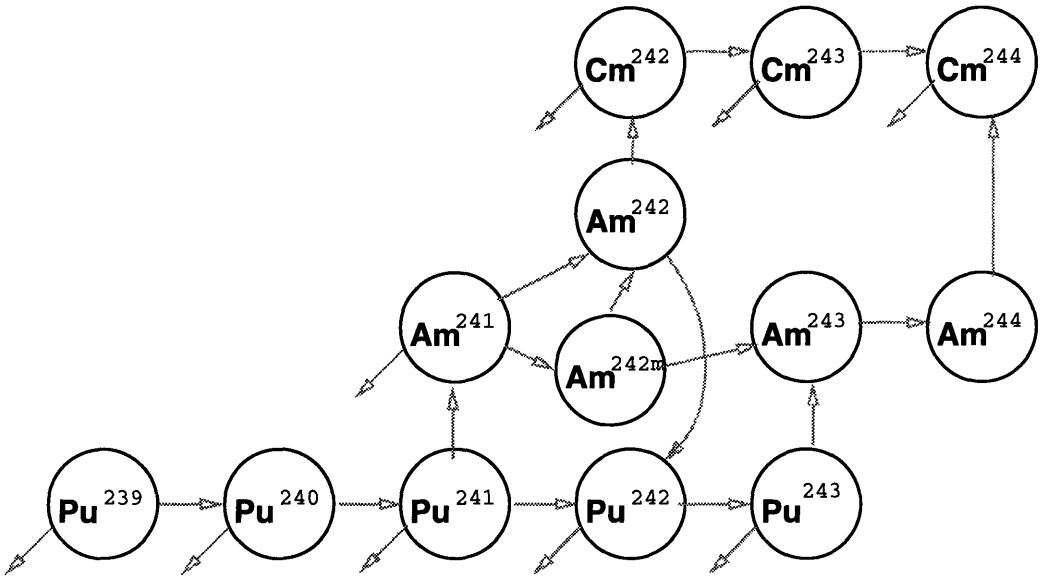

Figure 3-5: Transuranic nuclide chains producing Plutonium, Americium, and Curium.

the case of a solid lead target, that is being bombarded with an 800 MeV proton beam (as is the baseline ABC), the target yield is estimated to be 15 neutrons per proton for a lead target. The baseline ABC target design incorporates a target composite material made up of tungsten and lead. As part of the systems study in support of the ABC project, Krakowski [Krakowski, 1992] has developed an empirical expression for the neutron yield, Y, of the ABC target configuration as a function of accelerator beam energy, E_b:

$$Y = (E_b - E_0)/y \tag{3.13}$$

where y=30.1[MeV/n] and E_0=201.4 [MeV].

From this expression, knowing the current of the accelerator, an expression for source neutrons, S, in terms of target yield and beam current, I_b, can be written as:

$$S = YI_b/e \tag{3.14}$$

where e = 1.603 x 10^{-19} c (the charge of an electron).

For example, for an accelerator that delivers a 190mA (corresponding to 1.2x10^{18} protons per second) current at an energy level of 800 MeV, assuming a neutron yield of 20 neutrons per incident proton, the source provided to the slurry would be approximately 2.4x10^{20} neutrons per second.

Next, the neutronic character of the equilibrium slurry must be estimated. Most of what is known about the neutronic character of the ABC slurry fuel is taken directly from work supporting the ATW development effort. Although the slurry inventory of actinides and fission products for the ATW and ABC systems are likely to be different due to their functions, the neutronics behavior of the ATW can be used as a reasonable approximation for the ABC slurry. Davidson and Battat [DB, 1993] have calculated the equilibrium "lumped" average capture-to-fission ratio,α, neutron per fission, ν, and K_{eff} of the actinide slurry to be 1.6033, 3.0451, and .9245, respectively. However, since the capture-to-fission ratio is actually inversely related to flux level [SB, 1993], a range of capture-to-fission values considered, from 1.6 through 1.9, where 1.9 corresponds to a flux level of $10^{14}\frac{n}{cm^2 s}$.Finally, to be conservative in favor of a higher source demand, the lumped average fission yield, ν, for the actinide slurry (which contains Pu, Np, Am and Cm) will be assumed to be .29, or that of Pu^{239}.

With this information, the effect of the capture-to-fission characteristic of the slurry and total neutron leakage of the system on the source requirement can be calculated. Understanding this relationship is important for the following reason: If an accelerator-driven source is only required in designs where inventories of actinides and fission products are allowed to remain in the slurry, then the costs for developing this technology for sole purpose of processing plutonium may be questionable. As shown in Equation 3.2, this value is a function of the amount of non-fissile material (fission products and higher actinides) in the system.

From these assumptions, the required neutron source for various values of α and system neutron leakage were calculated, and the results are shown in Figure 3-6. As noted in Equation 3.2, the leakage term includes parasitic absorptions and the rate of leakage outside the system. For example, given a system leakage of 10%, as the value α is increased from 1.6 through 1.8, the source requirement increases from near zero (α =1.6) to $2.5\text{x}10^{19}\frac{n}{s}$ ($\alpha = 1.8$). Further, Figure 3-6 indicates that the neutron leakage factor does significantly impact source requirements. When the leakage increases by 10%, the source requirement increased by a factor of seven.

Next, consider how the capture-to-fission ratio and neutron leakage influence the corresponding accelerator current requirement. Figure 3-7 shows the relationship between the system leakage and capture-to-fission ratio for varying levels of accelerator current. As-

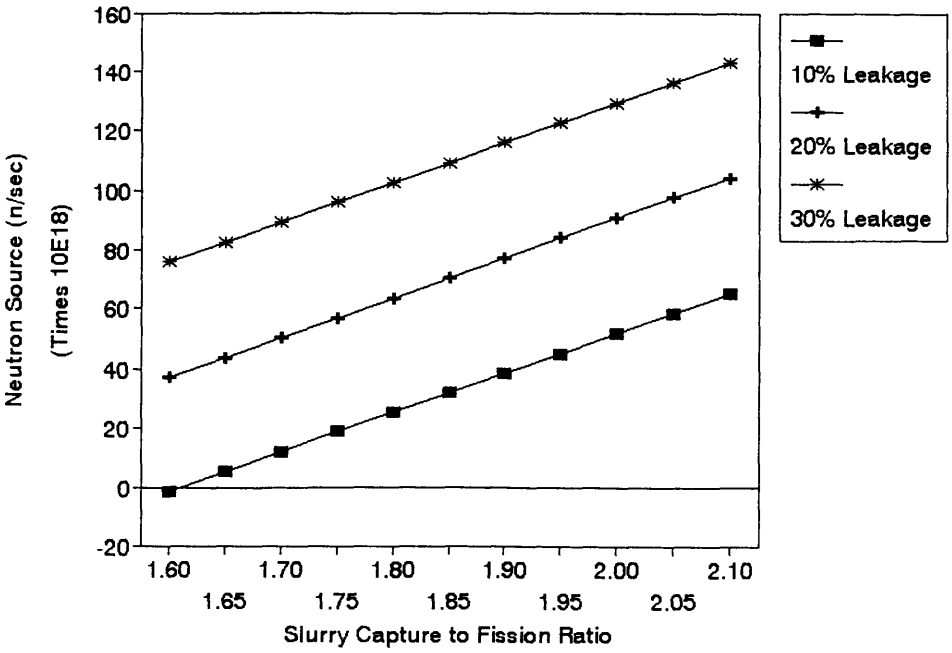

Figure 3-6: Calculated change in required neutron source due to changes in slurry actinide levels

suming the ABC baseline proton energy level of 800 MeV, a system with an α of 1.77 and neutron leakage of 10% will require the same accelerator current, 190mA, as a system with an α of only 1.61 and leakage of over 15%. Figure 3-7 also shows the increase in required accelerator current as amount of non-productive absorption material (actinides and fission products) is increased in the system, simulated in this example by an increase in leakage and α.

These results show that, for a given power, the required source neutron intensity increases as the relative amount of neutron capturing materials, such as higher actinides and fission products, build into the system. This implies that an ABC concept could conceivably be optimized for one of two purposes: (1) to maximize and control the amount of higher actinides and fission products in the slurry for the sake of transmuting them, or (2) to minimize the higher actinides and fission products in the system leaving a larger percentage of plutonium. Although the system function is the same in either case, the equilibrium amount of parasitic capturing materials will determine the source requirement for the system. As the graphs show, as you decrease the equilibrium amount of poisons in the system, that

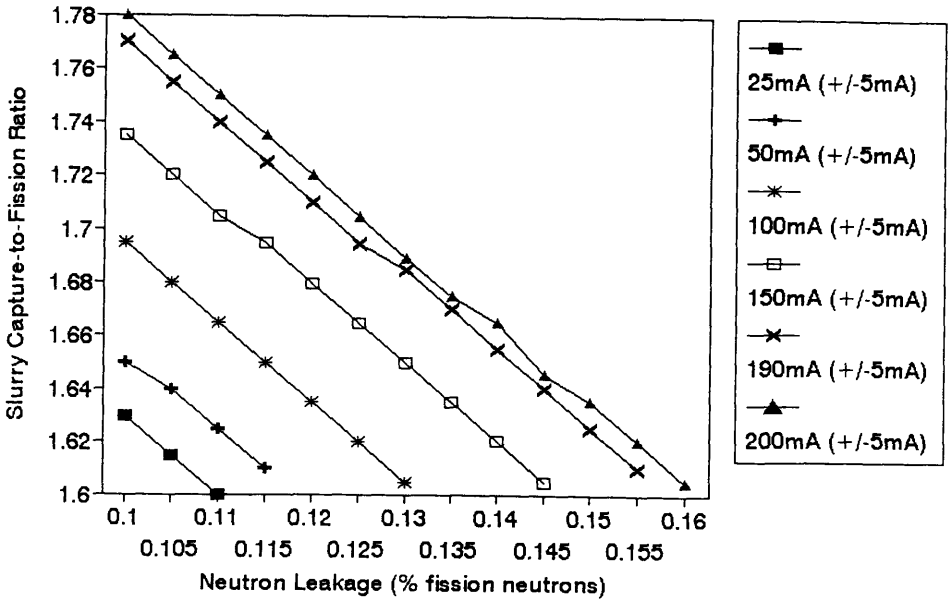

Figure 3-7: Relationship between α and system neutron leakage for constant current requirements

is, increase the thermal utilization factor, the source requirement decreases. As the source term decreases so, too, does the required current and energy level of the accelerator, thus, the cost and required development time of the accelerator component is reduced.

3.3.2 The ABC's Functional Requirements Defined

At the outset of this chapter, it was postulated that the most important and highest level requirement for plutonium disposition was to maximize proliferation resistance. In answer to this requirement, the generic concept of "fission reactor" was introduced as a possible solution. With a basic understanding of the ABC concept in hand, it would be helpful to describe it within the hierarchical design framework discussed earlier. Within this framework, the complex ABC system can be examined at the sub-system level. This helps gain an understanding of how the performance of each subcomponent influences the operation of the entire system, and how issues of technical maturity may influence the concept's successful development.

Figure 3-8 shows the hardware of the ABC concept mapped out in greater detail. At the

highest level, the fission reactor is chosen as the concept to satisfy the highest requirement for the WGPu problem (step 1), which is derived from need to create barriers against diversion of WGPu by terrorists groups for reuse nuclear weapons (Section 3.2).

At this point, the design begins to take form as the "physics" of the concept is considered. To define the concept at the second level of abstraction (step 2), the following question is asked: what functional requirements define the operation of a fission reactor (DP1)? As discussed in general terms in Section 3.3.1, there are three requirements: (1) a reactor must be capable of maintaining a neutron inventory sufficient to achieve the design fission rates or power level, (2) a reactor must provide a means of removing the thermal energy generated by fissioning, and (3) a reactor must provide a means of removing fission byproducts that accumulate in the fuel. Given these requirements, the second level concepts, which form the basis of the ABC system, can be described (step 3) as the solutions (hardware) that meet the three functional requirements, also described in Section 3.3.1.

To meet the first requirement, FR11, the ABC concept employs a subcritical reactor concept that maintains a steady state reactor-like neutron population without reaching a sustained fission chain reaction. Since this process operates at a neutron multiplication factor, K_{eff}, of less than one, the neutron population is kept constant by an large exogenous neutron source generated by an accelerator/spallation process. In this process, ions are accelerated to energy levels necessary to spallate with heavy metal target atoms. These inelastic collisions cause several nucleons, including neutrons, to be ejected from the target atom.

To meet the second requirement, FR12, the ABC concept proposes to use a D_2O based secondary cooling system similar to present CANDU reactor systems as the means of removing the thermal energy from the fissioning process. Using D_20 as the working fluid and moderator, this concept is able to cool the fuel via a primary slurry heat exchanger and convert the thermal energy into electrical energy with an efficiency of about 32%. It is important to note that this segment of the system is likely to be of primary concern to Russian policy makers who view the use of the weapons plutonium for energy production as an essential part of any disposition process.

To meet the third requirement, FR13, the ABC concept employs a continuous stream chemical reprocessing system that separates the desired transmutable materials from the parasitic absorbers. The baseline concept requires that the major actinides including pluto-

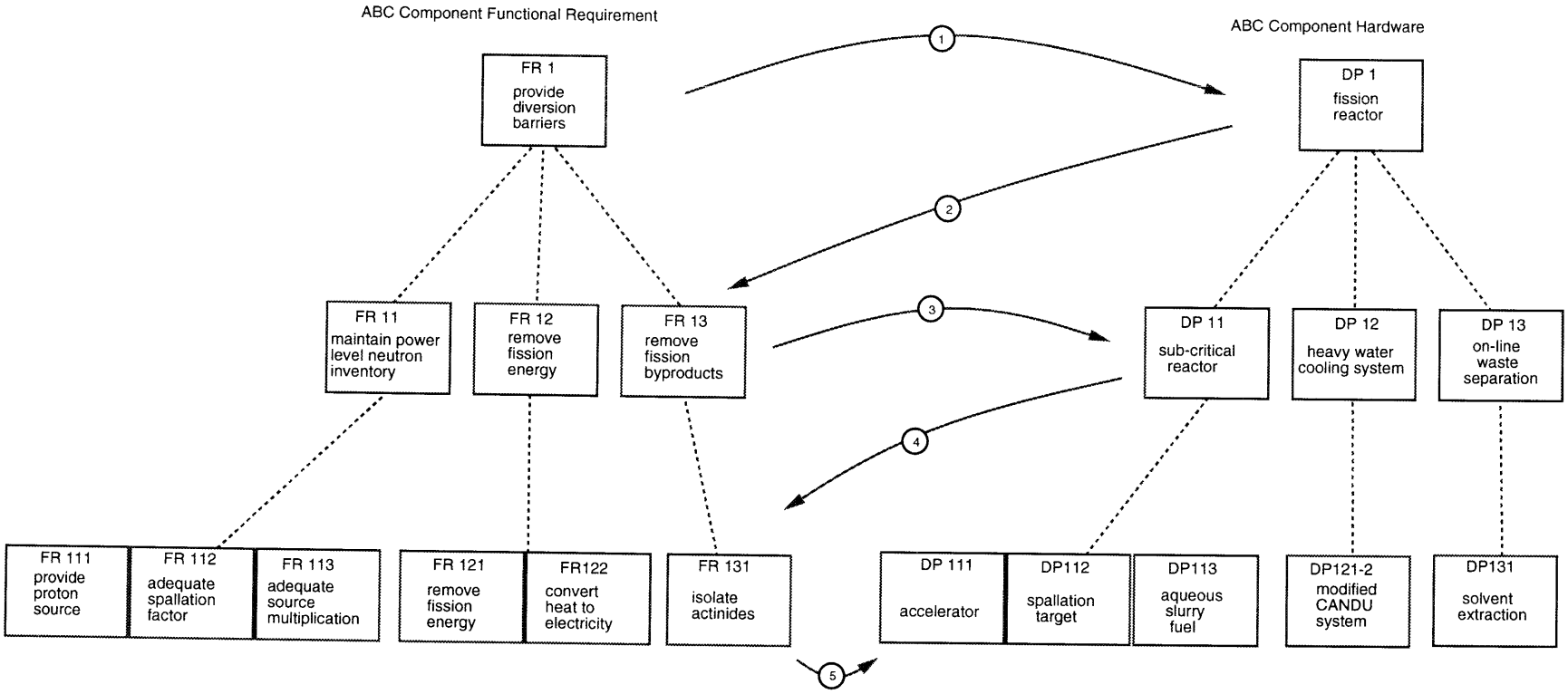

Figure 3-8: ABC FRs defined: (1) Fission reactor concept chosen to meet FR1,(2) Three level II requirements define for DP1,(3) Level II hardware concepts chosen to meet FR11, FR12, and FR13, (4) Level II hardware requirements defined, (5) Level III hardware concepts defined.

nium, neptunium, americium and curium be separated and reintroduced for further burning. In addition, key fission products will also be considered for possible transmutation.

Carrying the concept to the third level (step 4), a subcritical reactor has three basic requirements: (1) a proton source to initiate the spallation process, (2) spallation performance factor that maximizes the number of neutrons that are produced per incident proton, and (3), a slurry multiplication factor that is sufficiently subcritical to maintain safe operation, but near enough to critical to achieve design power levels, as discussed in the *Subcritical Nuclear Systems* Subsection of 3.3.1. As shown in the figure, the linear accelerator, spallation target concept, and the aqueous slurry concept define are the LANL proposed means to meet these requirements, respectively (step 5).

In this same manner, the CANDU secondary system meets the two requirements for the cooling system, which are: (1) remove fission energy and (2) convert heat to electrical energy. This second requirement is not essential but is added to meet the expectation that fission reactors will derive some benefit from the plutonium disposal process.

Finally, a fairly well established concept called solvent extraction will be employed to accomplish the requirement that the transmutable actinides, most importantly plutonium, be separated from the fission byproducts in order that they can be recycled for further transmutation.

In the next section, the details of each ABC component are discussed. The technical base for each system will be examined to determine how potential uncertainties may influence the performance of the ABC system.

3.4 Technology Base for ABC concept

3.4.1 High Current, Medium Energy RF Linear Accelerator

The advancement of linear accelerator technology resulting from the Strategic Defense Initiative (SDI), and related work elsewhere, has caused a resurgence in proposals to use accelerator-base technology for applications beyond particle physics research. Several areas have been considered, including breeder reactor systems, systems to produce tritium (APT), systems to transmute nuclear waste (ATW) and, most recently, as a basis to transmute WGPu. Although each application is unique, all require a linear accelerator with significant performance improvements over present technology.

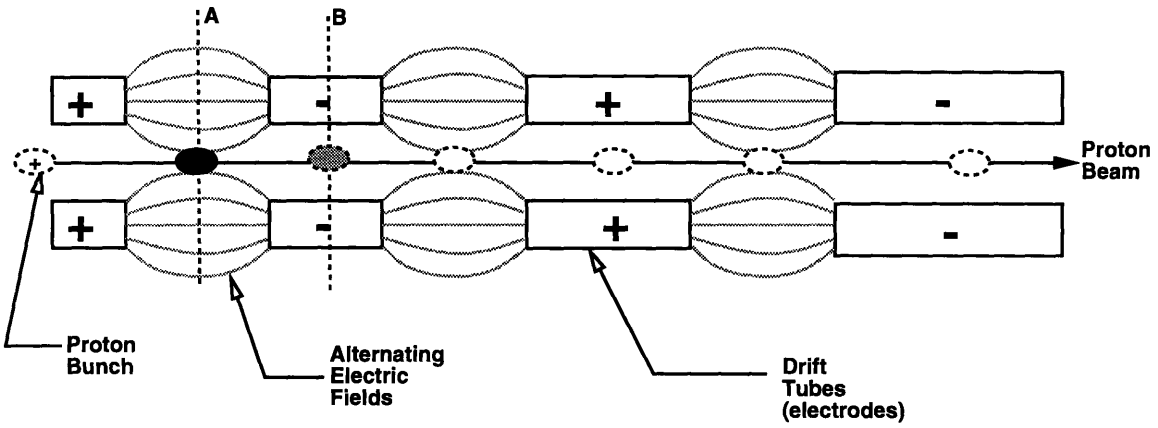

Figure 3-9: Basic Alverez accelerating structure configuration.

In simplest terms, the linear accelerator's function is to increase the kinetic energy of a charged ion, in this case a proton, by accelerating it in a straight line to velocities approaching the speed of light. High strength electric fields that exist between "gaps" in the accelerator path, force the positively charged particle to higher velocities as it travels along the accelerator, reaching, in some cases, energy levels of 1GeV ($1x10^9$ electron volts) or higher. An electron volt (eV) is equal to the amount of energy imparted on the proton through a potential difference of one volt. Radio frequency (rf) power supplies are used to drive alternating electric fields for the system. Although the basic accelerating concept is the same throughout a large scale accelerating scheme, specific structures must be designed for particular ranges of proton velocities.

The most basic accelerating structure used in modern day linear accelerators was developed in 194Os. The structure, known as the Alverez structure after its developer, is shown in Figure 3-9 and consists of a series of cylindrical tubes, called drift tubes, which are alternately connected to a high frequency oscillating power source. Protons are accelerated by electric fields in the gaps between the drift tubes that is generated by an rf power oscillator.

Because of the oscillatory nature of the rf power source - the electric fields alternate in direction once every cycle - the proton beam cannot exist as a continuous stream, but must travel in discrete bunches of protons. The analogy of "buckets" of particles is often used to describe the discrete nature of the proton stream. When the rf power cycle is in phase, or resonant, with the proton beam, the proton bunch is at location (A) which causes it to accelerate by some defined increment. When the phase of the rf power source is 180 degrees

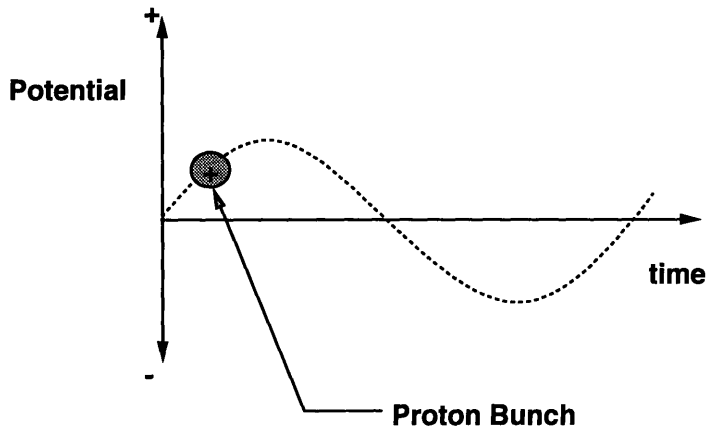

Figure 3-10: Resonant electric field cycle as a function of time.

from this position, the electric fields in the gaps have reversed, however, during this time, the proton bunch has moved into the drift tubes (point B) where the net electric field is zero [CM, 1991]. The proton bunch will "drift" to the next gap in time for the power source to once again provide a transverse force. Thus, only waves that are moving resonantly with the beam will produce a net acceleration. This concept is shown in Figure 3-10.

Given this, the length of an accelerating structure is a function of the rf power frequency. For a given accelerating structure, a single frequency is typically used. Thus, the drift tubes must be increase in length as the particle velocity increases so that the particle will arrive at the next gap in phase with the gap voltage [CM, 1991]. To decrease the size of a particular structure, it would be necessary to increase the frequency of the rf power source [CM, 1991, MGH, 1992].

Conventional linacs operate in pulsed mode because of the extremely high power demand from the rf power sources; however, there is no fundamental reason why a linear accelerator cannot be continuously driven by the rf generator, which corresponds to a 100% duty cycle (continuous wave (cw) operation). In this context, pulsed operation refers to the macroscopic pulses caused by turning the beam on and off, and is not referring to the pulsed (bunched) nature of the proton beam itself. The *duty cycle* for a linac can be defined as the ratio of time in operation to the time shut down. In fact, the pulse length can be adjusted for specific applications, and is limited only by the ability to cool the accelerator structure and cost [MGH, 1992].

As the duty factor increases, the amount of energy lost as heat energy in the walls of the

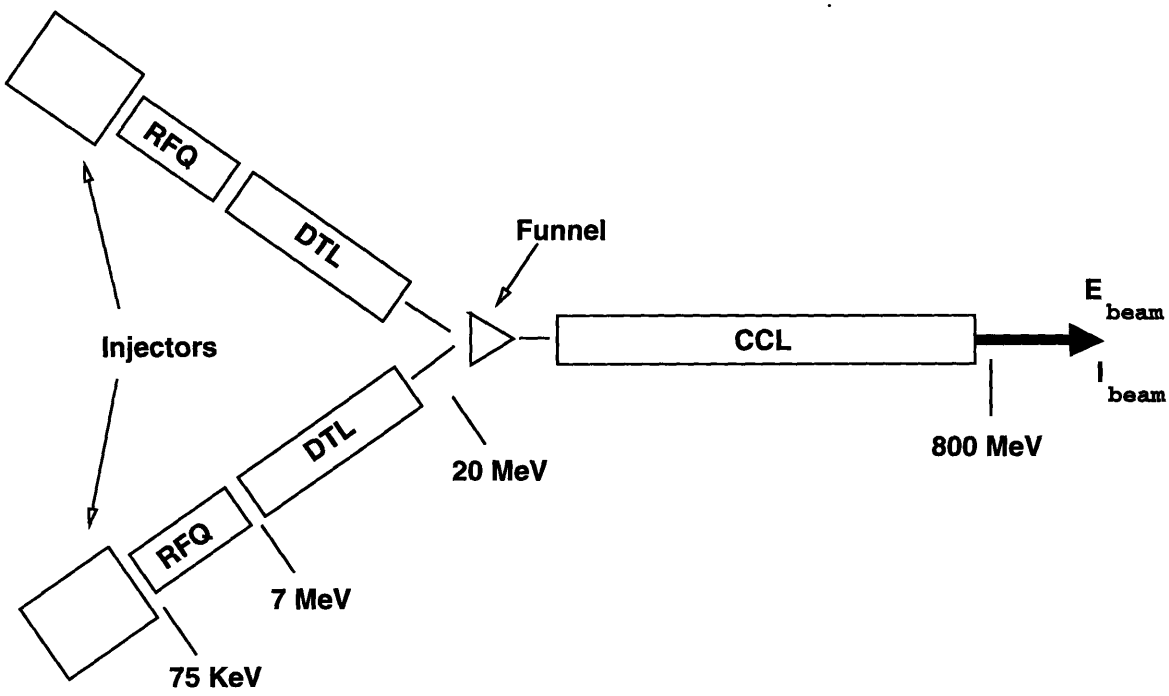

Figure 3-11: Linear Accelerator Configuration ABC System

accelerating structure greatly increases, and overall efficiency goes down. For high current systems such as the ABC, the heat generated by this mode is of critical concern.

ABC accelerator configuration

In simplest terms, the ABC accelerator system is designed to inject two 95mA proton beams into a main accelerating structure for acceleration to a nominal proton beam energy of 800 MeV and current of 190mA.

The ABC accelerator concept is based primarily on experience gained with the existing 800MeV pulsed accelerator located at LANL's Los Alamos Meson Physics Facility (LAMPF), and recent conceptual design work on the Accelerator Production of Tritium (APT) project. A fairly well developed conceptual design has been generated for the APT system which consists of beam launcher made from two DC injectors, two RFQs, two DTLs which are funneled into one beam at 20 MeV [LJS, 1992]. Figure 3-11 illustrates the present accelerator configuration for the ABC.

Injectors

The proton *injectors* provide a continuous proton beam that is both of sufficient current and high reliability. The base concept for this system specifies a total injector capacity of approximately 190mA, or 95mA per injector [SSS, 1993]. Protons are produced in the injectors by bombarding hydrogen gas with microwave radiation. The resulting reaction, which is done in a low vacuum chamber (10-15 torr), produces a proton plasma consisting of H^+, H^{2+}, and H^{3+} ions. These ions are pumped out of the system, by an electric field generated between two parallel 100KeV potential plates [Ste, 1993]. Thus, the energy of the protons leaving the injector are approximately 100KeV.

Frequently, an arch is generated between the two plates, momentarily causing the accelerating electric field to become zero. When this happens, the proton beam protons stops. Although this arching problem has merely been an inconvenience research applications, it may prove to be more serious for a system that requires continuous operation. The basis for the proposed injector design is derived from a resonance proton source presently operating at Canada's Chalk River Nuclear Laboratory (CRNL), which is capable of providing a continuous-wave 100mA current at an extraction voltage of 75 KeV.

Radio Frequency Quadropole

The Radio Frequency Quadropole (RFQ) is a new and revolutionary accelerator structure that was developed in the Soviet Union for low velocity ions acceleration [Jameson, 1991]. The RFQ structure has two functions. It is designed to convert the continuous proton stream ejected from the injectors into segregated *bunches* of protons, which is necessary for rf acceleration as discussed above, and accelerate the protons. In the RFQ, four electrodes run parallel to the beam axis for the entire RFQs length, and vary in charge and physical contour such that the proton beam is segregated into the appropriate proton bunch frequency to be introduction into the DTL [CM, 1991]. The ABC design concept specifies an RFQ acceleration of 75KeV (coming out of the injectors) to 2.5 MeV at a frequency of 400 MHz. As a result of SDI research, a cw ion source and RFQ at 100mA have been demonstrated at CRNL.

Drift Tube Linear Accelerator

The Drift Tube Linac (DTL) is designed to accelerate the proton beam from 2.5 MeV to approximately 20MeV in a fashion essentially the same fashion as discussed for the Alverez structure. This technology has been used at the LAMPF facility for several years and does not have outstanding limitations for application in an ABC configuration design [ERA, 1990].

Funnel

Achieving the high currents demanded for the ABC system poses a difficult challenge to the accelerator designer who is faced with limitations in injection currents [Wangler, 1993]. At low energy levels, the charge contained in each bucket of protons directly affects the beam's focus. As the proton density per bunch increases, in an effort to increase current, the beam focus degenerates, thus limiting the current achievable.

As a possible solution to this limitation, the ABC system proposes to combine two 95mA beams into one by a process called *funneling.* Funneling allows a significantly lower emittance (highly focused) beam at higher currents because, instead of increasing the proton density per bunch, it increases the number of rf buckets that are filled. In present accelerator designs, only about one fourth of the possible protons buckets are used. The funneling process would alternate the injection of proton bunches from each of the two low energy accelerators, thus spreading the rate of beam charge over more rf buckets. This, in turn, would reduce the effects of space charge and other intensity-dependent phenomena. If applied to the ABC system, it is estimated that the overall charge per bunch would be only be about 2.5 times greater than the present LAMPF facility, which is within an experience base that is well understood [LJS, 1992].

The concept of combining two lower current proton beams into one high current beam is a new and yet unproven concept. Preliminary work as part of the LANL SDI program has demonstrated the basic principles of funneling. In one experiment, the effect of passing a 60mA, 5MeV current through the funnel structure proved successful [DBB$^+$, 1992]. However, significant development work is still necessary to verify the process alternately [Jameson, 1991].

Coupled-Cavity Linear Accelerator

For energy levels above 200MeV, the efficiency of the Alverez structure decreases [MGH, 1992], thus an rf accelerating structure for higher velocities is necessary. The coupled-cavity linac (CCL), which is presently being used in the LAMPF facility, is made up of a series of resonant cavities that, when coupled together, accelerate the protons to energy levels of 800 MeV and higher. Since a majority of the overall length of the linac is comprised of the CCL structures, the CCL constitutes a major portion of the accelerator cost [Jameson, 1983]. The CCL section length determines the final energy of the beam, thus, the energy level that can be achieved is primarily a function of length. For the ABC concept design, after a DTL has increased the beam energy to 100 MeV, a CCL is used to increase energy of the now combined 190mA beam to 800 MeV.

Radio Frequency Power Source

The radio frequency power that drives the accelerating structures originates from an electron-beam high-frequency fr generator called a klystron [MGH, 1992]. The klystron, which can produce very high rf power in frequencies from a few MHz to several tens of GHzs, is coupled, via an inductive loop, to a wave guide that pipes the power to the accelerating structure [CM, 1991].

The rf power source for the baseline RFQs and DTL configuration will provided by existing commercial available, 1 MW cw 350 MHz klystrons. However, the CCL will require several 700 MHz, 2MW rf power supplies to drive the high current, medium energy proton beam [WB, 1992], which are considered immature technologies [APT, 1993], and unavailable for production. The high frequency is necessary because the frequency (or rate) of proton bunches has doubled due to the "funneling" of the two lower current beams.

Technology base for ABC linac

Table 3.1 list several research linear accelerators and their corresponding energy level, current, and availability [Kurokawa, 1989]. For research applications, linac technology can be said to be mature given the extensive experience in high energy, low current, beams. However, for applications in which continuous wave and high current systems are needed, no full scale experience is available. Some recent improvements in RFQs close the gap towards

Accelerator Technology						
Title	When	Type	Current (mA)	Energy (MeV)	Beam Type	Availability (%)
CERN	1959	linac H+	7.0	50.0	pulsed	-
IHEP	1967	linac H+	100.0	100.0	pulsed	99 *
CEN	1969	linac H+	20.0	20.0	pulsed	98 *
BNL	1970	linac H-	25.0	200.0	pulsed	95 *
Fermilab	1970	linac H-	35.0	201.0	pulsed	97 *
LAMPF	1972	linac H+	17.0	800.0	pulsed	85 *
KEK	1974	linac H+	10.0	40.3	pulsed	95 *
CERN	1978	linac H+	140	50.0	pulsed	99 *
FMIT*	1982	linac-RFQ H+	50.0	2.0	continuous	-
Rutherford	1983	linac H-	14	70.4	pulsed	-
CRNL	1988	linac-RFQ H+	50.0	0.6	continuous	-
ABC	tbd	linac H+	150.0	800.0	continuous	-
* data given by LANL						

Table 3.1: Select proposed and active linear ion accelerators

this end.

Figure 3-12 shows a constant source requirement curve of $3.3\text{x}10^{19}\frac{n}{s}$ which is taken from an example case for the ATW conceptual design [LJS, 1992]. Using the empirical model represented by Equation 3.13, beam current requirements were calculated for a range of energy levels between 0 MeV through 800 Mev. The capacity of existing technologies are plotted against this goal to determine how far present experience is from this capability. As noted earlier, much of the linear accelerator experience base lies in low current systems.

From the figure, it appears that the accelerator facility that lies closest to the proposed ABC system is the LAMPF facility at LANL, which has a beam energy and current of 800MeV and 17mA, respectively. Since beam energy is primarily a function of accelerator length, the main obstacle to achieving an ABC like proton beam character lies in the difficulties related to high current capacity.

Recent Accelerator Technology Reviews

Two extensive technical evaluations have been conducted for a 1GeV and 200mA accelerator concept as part of the Accelerator Production of Tritium (APT) program, which provide a convenient envelope within which to comment on the maturity of the slightly less ambitious 800MeV-190mA ABC proposal.

The Energy Research Advisory Board (ERAB), in a report dated February 7, 1990, published their evaluation of a 1.6GeV, 250mA linear accelerator-based concept to produce tritium [ERA, 1990]. They note the lack of experience in using linac technology to produce

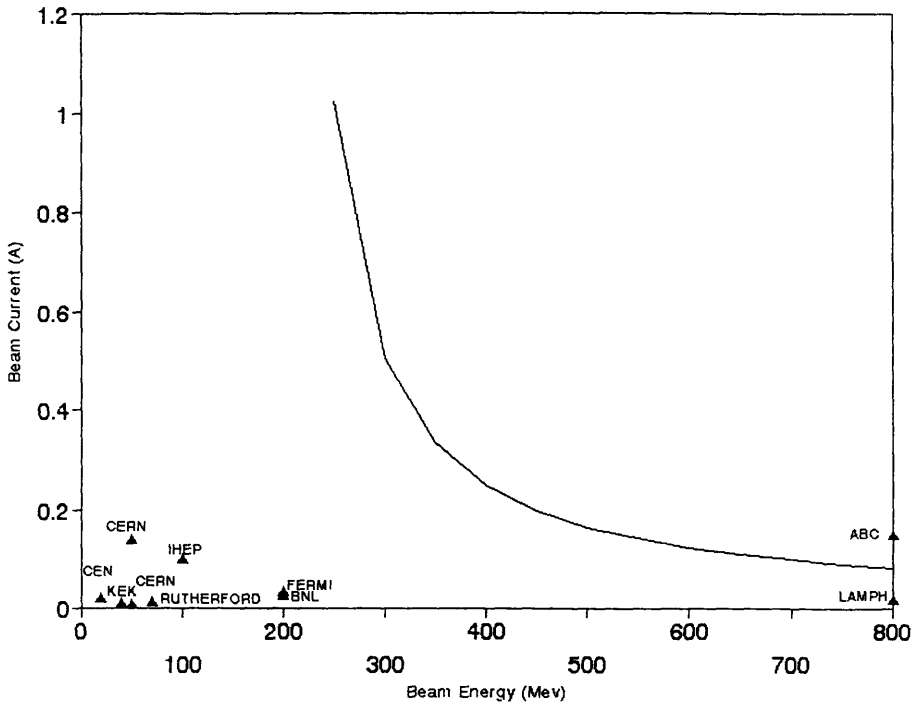

Figure 3-12: Source requirement for various technologies

neutrons at the rates proposed. The availability required for a neutron production machine, which must run continuously at full design loads, would be much higher than current linac technology. They summarize the technology this way:

> The continuous-wave RF linear accelerator approach is technically sound. However, the lack of accelerator experience at APT conditions, a target system that is only conceptual, and the need for component development and overall APT system testing and evaluation, prevent the immediate translation of the technology...

The JASON panel, in their report dated January 1992, cite similar findings [DBB+, 1992]. In their evaluation, they considered a slightly less ambitious 1.6GeV and 125mA proton beam design goal. Citing no significant engineering issues, the panel concluded that:

> In agreement with the ERAB panel, this Panel believes that there exists an adequate base of experience and technology to give confidence that a successful APT accelerator can be built and operated with adequate availability and reliability.

It would be reasonable to conclude, then, that the basis for the ABC linear accelerator concept is sound. This conclusion only holds for accelerator technology in general; however,

issues specifically related to the reliability requirement for a systems that is producing power must also be considered.

3.4.2 Tungsten-Lead Spallation Target

Traditionally, spallation neutron sources have been used for material scattering experiments in research laboratories; however, as early as 1948 the possibility of generating large neutron yields from materials excited to high energy states was recognized for its application in energy related systems, such as breeding fissile material [FB, 1983]. However, these ideas have mostly remained as paper studies because the accelerator technology necessary to produce the required ion currents for such applications was well beyond reach. Within the bounds of accelerator capability, extensive development work to optimize the neutron yield of the spallation target has been done and is well documented. However, as accelerators increase in ion intensity, the requirements for spallation targets also increase, thus, spallation targets must be designed and tested in parallel with accelerator development [Beard, 1993].

The principal function of the spallation system, which includes a heavy-metal spallation target and moderator, is to convert a high intensity, high energy proton beam into neutrons and deliver them to the blanket slurry fuel to be used in useful neutron reactions. There are two design goals for the spallation target system. First, the target spallation system must be designed to maximize the neutron production, which is a function of target material used and the energy of incident proton beam. Second, the target must be designed to maximize the neutron leakage out of the target to allow them to be used in "useful" neutron interactions in the slurry [Arthur, 1993]. Give these goals, the two primary system characteristics must be considered in a particular target design are (1) neutron yield per incident proton, and (2) power density of the target system. The power density is of particular interest because the proposed increase in accelerator current for medium energy ranges (approx. 800MeV) greatly increases the disposition power to the target, an order of magnitude higher than present spallation target experience.

The spallation process

The term "to spallate" means to chisel a stone. In physics, spallation means to break off neutrons, protons, and other subatomic particles from the nucleus of a heavy atom by bombarding it with an medium energy particle (approximately 1000MeV). In a spallation

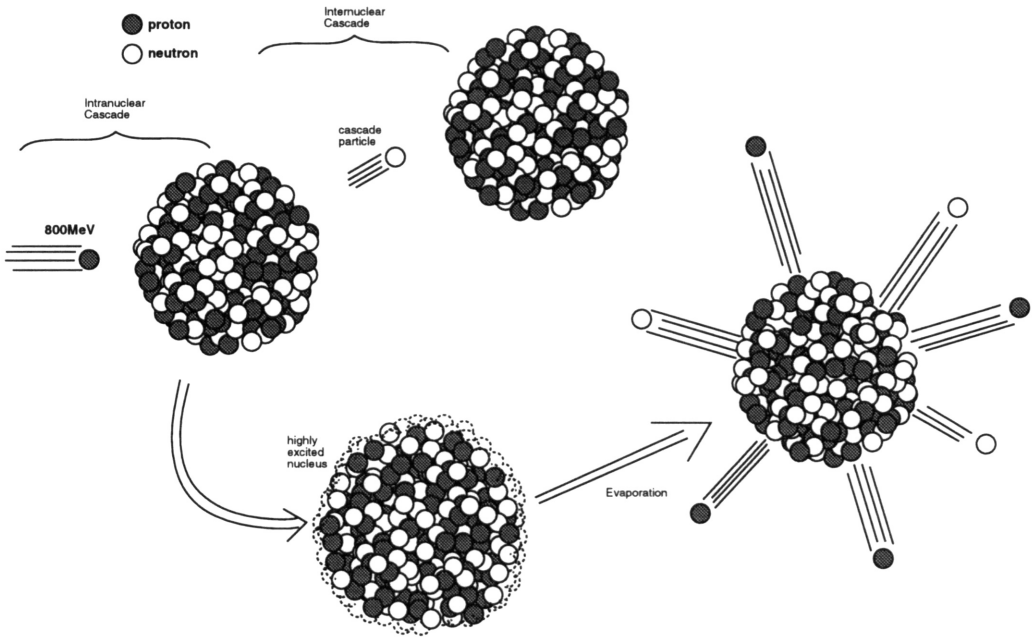

Figure 3-13: The spallation process occurs in two stages: intranuclear cascade and evaporation.

reaction, the nucleons are thought to emerge in a two-stage process beginning with a reaction called an "intranuclear cascade", between the incident proton and the target atom, and followed by the ejection of nucleons through an "evaporation" process as the excited target atom sheds excitation energy [CFH, 1982].

Figure 3-13 illustrates the two-step process. In the first stage, the medium energy incident proton collides with the heavy metal target atom. The proton imparts energy to individual nucleons inside the atom which result in further collisions, giving rise to particle "cascade" within the atom. In a very short time, on the order of 10^{-22} seconds, nucleons are ejected from the nuclei. The ejected nucleons are then free to interact with other target atoms, which produce further cascades (internuclear cascades) [Bauer, 1982].

A portion of the original kinetic energy imparted to the target atom during the intranuclear process does not leave in the form of direct particle ejection, but remains, causing atom to remain in an excited state. On a time scale much longer than the original spallation event, approximately $10^{-17 to -18}$seconds, the atom begins to shed this excitation energy in the form low energy nucleons [CFH, 1982]. This process is called "evaporation".

The total number of neutrons that are produced from the spallation process depends on the type of target material used and the energy of the incident proton. The target material is important because the spallation cross section and the fissioning characteristic of the material directly affect the number and type of interactions that take place. For example, for a target that uses U-238, neutrons are produced by the spallation process, as described above, and by fissioning.

Early experiments to determine the spallation yield for various materials showed that the number of neutrons that emerge from the spallation process was linearly related to the energy of the proton beam [Bauer, 1982]. The materials most commonly used in designing spallation targets are Ta, W, Pb, Bi, Pb-56, w/o Bi, Th, U, and U-238, and of these, U-238 provides the largest neutron yield (because of fissioning) and Ta the smallest [FB, 1983].

Considering the target system on a macroscopic level, a portion of the total energy contained in the form of kinetic energy of the proton is translated into neutrons that travel outside the target region; however, much of the kinetic energy of the proton beam is translated directly thermal energy which is contained in the target material itself. The charge particles from the evaporation process, because they are of relatively low energies, and the recoiling residual nuclei, result in localized energy disposition normally a few tens MeV near the spallation site [CFH, 1982]. For a given proton beam penetration, the corresponding heat intensity decreases exponentially as it progresses through the target material [Stelzer, 1982]. As an example, for a beam energy of 800MeV (baseline ABC) and a current of 150mA, the power deposited on the target system is approximately 120 MW.

ABC Target Concept

Figure 3-14 illustrates the base target design. Two primary concerns have driven this concept: (1) maximize generation of useful neutron per incident proton, and (2) ability to withstand the extreme heat energy disposition imposed by the high current, medium energy proton beam. The proposed target station will use a composite spallation material configuration with tungsten as the primary axial spallation material and lead to surround the target for radial coverage. For the beam stop, Zircaloy-4 is proposed. In the configuration shown, calculations indicate that about 47% of the low energy neutron production will occur from tungsten spallation interaction, 45% for lead, and only about 7% for the Zircaloy-4 beam catcher. However, depending on the specific design, the percentage of neutrons

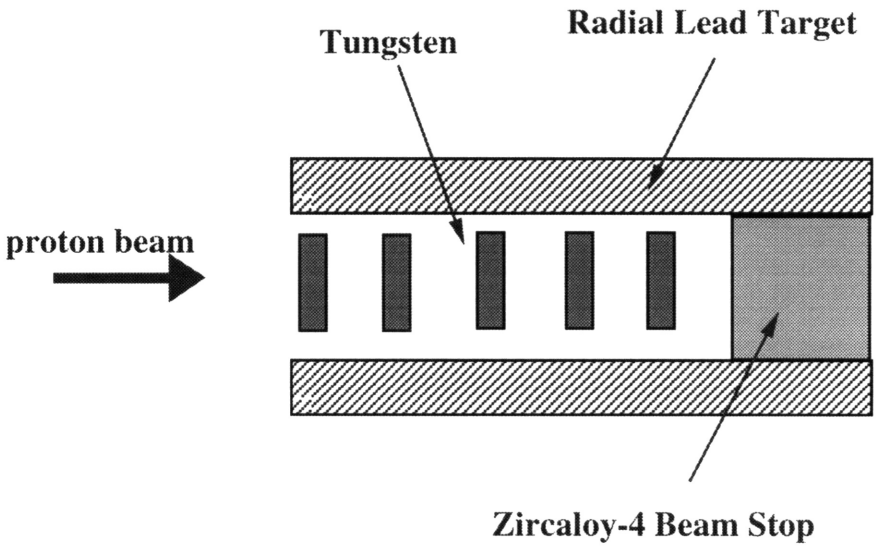

Figure 3-14: Base target design

produced by the lead material can vary anywhere from 30% to 50% [Beard, 1993]. The target yield is expected to be 18 neutrons per incident proton for a beam energy of 800 MeV [ADL, 1992].

Tungsten was chosen as the primary spallation material mainly because of its large heat capacity relative to that of lead. Assuming a beam energy of 1.6 GeV and 200mA, the highest beam power density will occur in the tungsten region with a magnitude of 1.992 MW/l [DOE, 1993a]. The ERAB panel [ERA, 1990] notes that the power densities for the ATW target (1.6GeV, 200mA) will be similar in power density to that experienced in light water reactors. As a comparison with other types of reactors, a PWR core has a power density of .105MW/l, and an LMR reactor has a corresponding value of .280MW/l [TK, 1990].

The maximum temperature of the tungsten is calculated to be 159 ^{o}C, which is well below its corresponding melting point of 3410 ^{o}C [APT, 1993]. The peak temperature for the lead material is calculated to be 102 ^{o}C, which is reasonably above its melting point of 327^{o}C [APT, 1993]. The tungsten inner region will be cooled with heavy water, which serves as the primary target coolant and moderator [ADL, 1992]. The secondary working fluid will be light water. The energy will be dispersed into the environment using a standard cooling tower [CISK, 1992].

Target geometry and material selection are the two variables in control of the target

designer to maximize the number of useful neutrons that are generated per spallation proton. One disadvantage of using tungsten as the primary target is its large characteristic neutron capture cross section [Beard, 1993]. Particularly for large diameter tungsten targets, parasitic absorption becomes a significant factor in overall neutron yield. To mitigate this effect, the tungsten target material, which is in the form of bundled rods, is placed in the forward section of the target configuration with gaps between them. It is believed that this spit target design will allow neutrons to escape into the moderator before they can be parasitically captured by the tungsten atoms [Arthur, 1993]. Lead, on the other hand, has a low neutron absorption cross section, thus, serves as an effective secondary "catcher" for the protons that did not interact with the tungsten material.

Spallation Target Technology Base

Target yield behavior as a function of proton energy level is fairly well documented and, based on target station design and research to date, there is little question that the target yield of 18 neutrons per incident proton can be achieved for an 800 MeV proton beam. Fraser and Bartholomew [FB, 1983] reference an early material yield experiment conducted jointly between the Chalk River Nuclear Laboratories and Oak Ridge National Laboratory in the late 1960s that seems to validate the estimated yield numbers provided for the ABC target design. In this materials study, the neutron yield was measured for several target materials for a range of proton beam energy between 500MeV to 1500MeV. Figure 3-15 shows results of the yield material study (figure taken from [FB, 1983]). The targets used in the experiment were cylindrical in shape, and the first dimension in the figure is the diameter of the test target and the second dimension is the length.

For this discussion, it is important to note that the magnitudes of neutrons yields for the base materials are within the same order as the target proposed for the 800MeV ABC spallation system. For example, if it were possible to employ totally lead target, the yield for this energy level would be approximately 15 neutrons per incident proton, for the 20.3 cm diameter target. Fraser further notes that there is proportional relationship between neutron yield per incident proton and atomic mass number. Thus, employing a tungsten target, with a yield curve that lies between Sn and Pb, would also be within the 10 to 20 neutrons per proton range. In an extreme case, a design could employ a uranium target which would achieve much higher yields than its counterparts. For an 800 MeV proton,

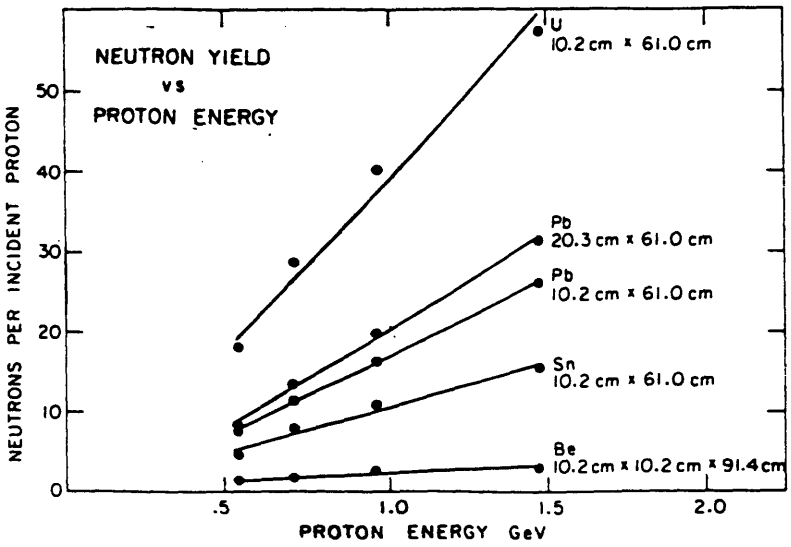

Figure 3-15: Neutron yield for varying proton beam energy levels.

according the figure, a uranium target would yield nearly 30 neutrons per incident proton. Uranium is an example of a spallation material that exhibits both fission and spallation behavior, which causes the overall neutron generation to be greater [FB, 1983].

The information provided by the spallation yield study noted above has been the basis for spallation target design. Cloth, Filges and Hecker [CFH, 1982], for example, present calculations for the DIANE target station design which is based on a rotating wheel target concept. In their analysis, they assume that a pulsed 1100 MeV proton beam impinges on an aluminum-clad lead target, that is moderated with D_2. With this configuration, they calculate that the neutron yield will reach 25.3 neutrons per incident proton.

Although the possibility of achieving the ABC target yield can be stated with some degree of certainty, the same is not true for the ability of the target receive the large beam power deposited during the spallation process. Because of the dramatic increase in beam current from present day research spallation facilities, the target power for the ABC and other proposed *production systems* is at least an order of magnitude larger then any target station presently operating or planned for the near future. Table 3.2 lists the beam characteristics and target power levels for several existing and proposed spallation facilities (data taken from [FB, 1983]).

It is important to remember that spallation source technology has thus far been driven by research demands which do not necessarily required large steady neutron sources. Therefore, beam power has remained in the kW to 1MW range. For example, for the high energy LAMPF facility, total target power has been in the range of 13kW for a rotating type

Facility	Location	Type	Operating Energy (MeV)	Current (mA)	Status	Target-Power (MW)
Harwell	UK	pulsed	150	.0096	shut down	-
Nevis	US	pulsed	385	.0012	shut down	small
WNR	US	pulsed	800	$\leq$ .02	operating	.006
WNR+PSR	US	pulsed	800	7.1	-	-
KENS	JAPAN	pulsed	500	.0019	operating	.002
IPNS I	US	pulsed	500	.012	completed	.009
IPNS II	US	pulsed	800	.48	proposed	.5
SNS	UK	pulsed	800	.2	-	.23
TRIUMF	CANADA	cw	500	$\leq$ .1	operating	.04
SIN	SWITZ	cw	600	2	planned	.4
SNQ	FRG	-	1100	5	proposed	3.3
ING	CANADA	cw	1000	65	aborted	38
LLL	US	cw	500	.375	study	110
ANL	US	cw	1000	.3	study	170
LAFR,LAFP	US	cw	1500	.3	study	-
ABACS	US	cw	1000	.3	study	190
CRNL	CANADA	cw	1000	.3	study	450

Table 3.2: Active and Proposed Accelerator Target Station Power Levels

target, to 600 kW for isotope production and beams stops [WSS$^+$, 1992]. The highest power target planned is the SINQ cw machine with a 1.2 MW power target.

The high energy, continuous wave machines listed above have been proposed for energy production systems. Although studies do indicate that the power densities in these targets are comparable to reactor systems, designing the equivalent in spallation targets poses a significant challenge. Fraser *et al.* [FB, 1983] concludes that:

> [I]t is quite conceivable that the optimum practical design [target design] will be determined more by engineering considerations of heat removal, material compatibility and structural integrity under irradiation than by any requirement to achieve the best possible yield.

This conclusion was echoed recently by the JASON panel in their evaluation of the 1.6 GeV, 120mA, 192MW target station where they note that "considerable engineering and development would be necessary to provide appropriate fluid and heat transfer systems" [DBB$^+$, 1992].

3.4.3 Actinide and Fission-Product Separation and Reprocessing

The basic concept by which actinides and other fission byproducts are chemically separated was actually established over a century ago. The solvent extraction process, which is the basis for the ABC processing system, was first applied in 1841 when it was discovered that the solubility of uranyl nitrate in diethyl ether coupled with the ability of ether to extract uranyl

nitrate from aqueous nitric acid could be used to separate uranium [Nelson, 1985]. Since then, solvent extraction has been used for separating and purifying plutonium, uranium, and other actinides in various quantities and applications. The demand for large quantities of fissile material by the defense and commercial sectors required that high capacity separation processes (tons per year) be developed for plutonium and uranium. However, processes for higher actinides such as americium and curium were developed primarily to meet a much smaller market demand in research. Thus, production capacities for these materials remains at the grams per year level. However, in recent years, as attention focuses on the problem of accumulating nuclear waste, the possibility of chemically separating and transmuting the higher actinides and some fission products as an alternative to long term storage for some types of nuclear waste have caused many countries to invest a substantial research effort toward this end[Newman, 1991]. It is within this new "market" that the ABC concept proposes to develop the required actinide separation process technology.

The primary function of the ABC chemical processing system is to separate higher actinides, most importantly plutonium, and key fission products. By nature of its slurry-based fuel system, the ABC requires that the actinides be separated quickly to maintain the desired neutronics within the "core" region. This process has a two-fold purpose. Chemical separation provides the primary mechanism for controlling reactivity (maintaining a constant K_{eff}) in the system by *removing* the materials that cause changes in core reactivity. This is unlike solid fueled systems which control reactivity fluctuation by inserting or removing control rods. Secondly, the separation process provides a means by which plutonium can be continually recirculated for extensive or complete burnup. This is also true for any material recirculated to the core region.

Baseline ABC Actinide Separation Process

Figure 3-16 illustrates the basic solvent extraction process. The desired species, which is in an aqueous solution, is placed in contact with an organic extractant that, because of a lower density, remains above it. The mechanism of separation for a particular material is a function of the acidity of the aqueous solution, thus, by varying the pH of the aqueous solution, specific actinides and fission products within the aqueous phase can be drawn into the organic phase. The remaining constituents in the aqueous solution can be further processed down stream [Cleveland, 1979].

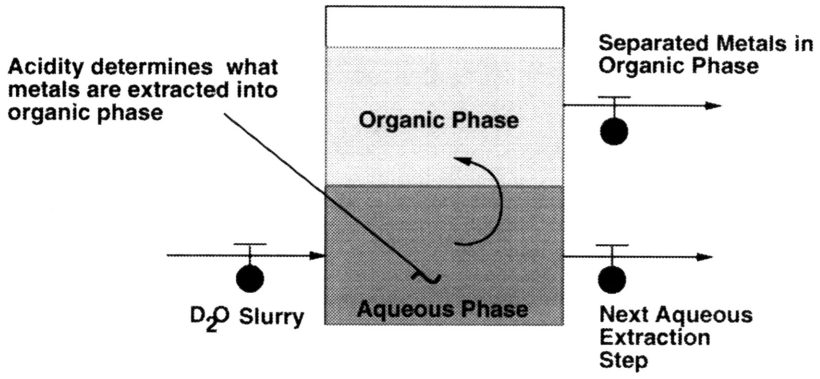

Figure 3-16: Basic solvent extraction process

Because americium and curium fission more slowly than plutonium and neptunium and are also more difficult to separate from the fission products, the ABC concept separates the Np/Pu and the Am/Cm recirculating loops. Thus, the chemical process stream for the ABC system will consist of two main process steps. First, the plutonium and neptunium, will be separated using a variation on a well established PUREX solvent extraction process. Second, the americium and curium, will be isolated from the lanthanides using the TALSPEAK process [ADL, 1992]. The general schematic for the ABC flow sheet is illustrated in Figure 3-17.

Dewey *et al.* [DJM$^+$, 1993] describe the proposed ABC chemical slip-stream process. The plutonium oxide slurry will have a residence time in the transmuting core region of about 15 days before it is removed for processing at a rate of about 7% of the total slurry flow per day. An equal 15-day cooling period for the slip-stream will be employed before the first processing stage begins. After the plutonium-neptunium extraction, the aqueous stream containing americium, curium and fission products will be stored for further cooling, for a total of total of 90 days, before the trivalent actinides are recovered. This additional cooling is necessary to allow short-lived highly-active fission products to decay to a level which can be tolerated by the trivalent actinide extraction process, and, in turn, allowing a 90-day average residence time for these actinides in the primary loop.

After the designated initial cooling period for the slip stream, the D_20 is evaporated from the slurry mixture, and the concentrated oxide slurry is dissolved in HNO_3 and a quaternary ammonium anion exchanger, Aliquat 366 (methyl tricaproylammonium chloride). Aliquat 366 is becoming increasingly important as a substitute for the tributyle phosphate-hydrocarbin (TBP) extractant which is used in PUREX process because it is believe to be

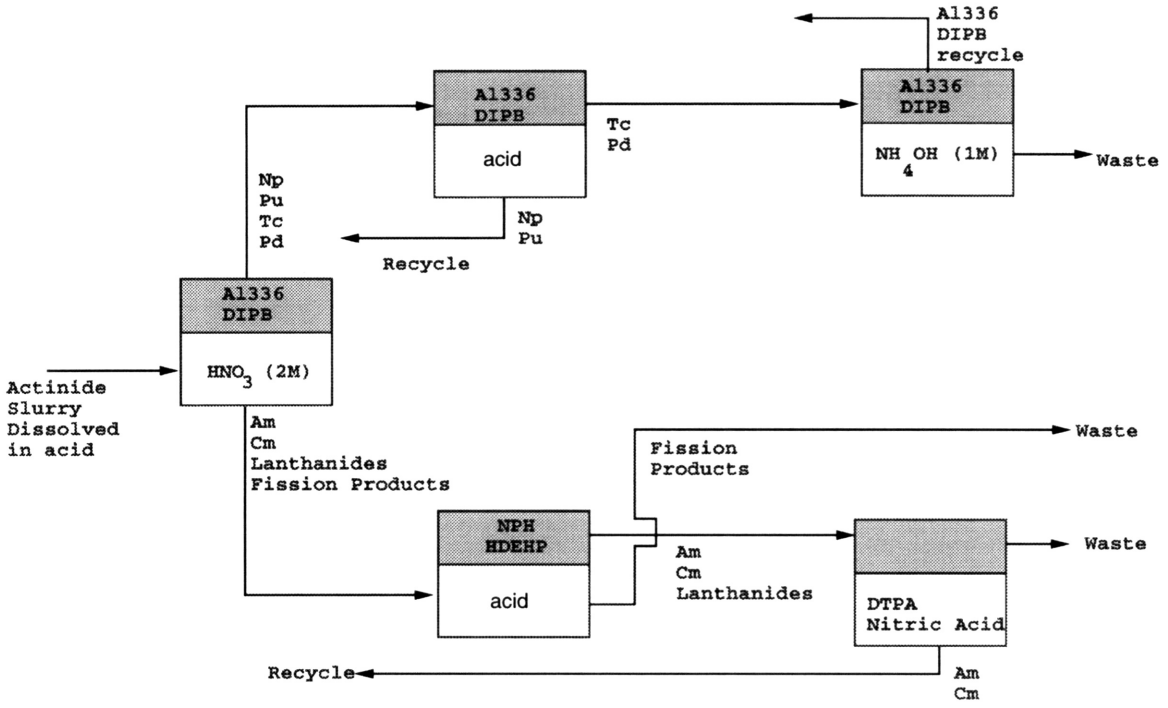

Figure 3-17: Actinide/Waste Separation Process

more stable in high radiation environments such as in the ABC system. Next, Pu and Np are back-extracted with 0.1M HNO_3, leaving Tc and Pd in the organic phase. In a similar fashion, the Tc and Pd are extracted and eventually stored as waste. Finally, a controlled thermal denitration process is used on the plutonium-neptunium laden aqueous strip solution to produce the mixed-oxide recycle, NpO_2 and PuO_2, which are then reintroduced to the main slurry loop.

The remaining aqueous solution is cooled for 90 days before the americium, curium and lanthanides are separated from the rest of the fission products using the TALSPEAK process. The TALSPEAK process, which was develop in 1964 [SM, 1980], coextracts the trivalent actinides and lanthanides (rare earths) into 1M HDEHP (di-2-ethylhexylphosphoric acid) from an acid solution in a similar fashion as discussed above for plutonium. Once the Am, Cm and lanthanides are loaded into the HDEHP organic phase, the actinides are back-extracted and separated from the rare earths with an aqueous phase containing the DTPA (diethylenetriaminepentaacetic acid). As before, oxide forms of the actinides are produced and recycled into the main slurry loop, while the lanthanide fission products are

treated for permanent disposal on-site. Similar trivalent actinide separation processes are discussed by Carley-Macauly [CM, 1985] and Horwitz and Schulz [HS, 1985].

Actinide Processing Experience

Today, there is no facility or process that has been demonstrated at the capacity or activity level required for the ABC on-line processing, however, aqueous based separation technologies have been defined and verified under less extreme conditions. In fact, it was the need to estimate a complete and reasonably verifiable material balance that guided ABC developers to choose an aqueous based slurry fuel for their concept [DJM$^+$, 1993]. As noted earlier, the ABC concept has two technology "pools" from which to draw experience: (1) processes developed for separating plutonium and uranium for military and commercial fuel applications, and (2) processes developed to supply higher actinides for research needs. However, the latter is most relevant for the ABC because americium and curium separation at high radiation levels will be required.

The ABC process is based primarily on the experience and technologies developed for the Transuranium Processing Plant (TRU) at Oak Ridge National Laboratory [KBC, 1981], where the separation of transuranic materials is conducted at high radiation levels comparable with the ABC system. The TRU facility is the production center of heavy elements, for research throughout the country, and since beginning operation in 1966, has produced transputonium elements in the gram to kilogram levels for many heavy-elements from plutonium through fermium. The characteristicly high radioactive nature of these transplutonium materials require remote handling. The TRU facility is made up of nine heavily shielded hot cells in which solvent extraction, ion exchange, and precipitation operations are conducted.

Other facilities similar to the TRU have been proposed or are operating throughout the world, most notably in France and Germany. A summary of Germany's development work can be found by Koch and Stoll [KS, 1981], while Koehly *et al.* [GBM$^+$, 1981] present the transplutonium separation technology developed in France.

Separation Processes in High Radiation Environments

Although a significant increase in separation production level is required for the ABC processing system, radiation damage concerns dominate the uncertainty of the proposed pro-

cess. The short cooling times before processing, 15 days for plutonium separation and 90 days for trivalent actinides, translate into extreme radiation and thermal loads which greatly limit the choice of solvents and chemical reagents that can be used, and pushes the actinide separation technology beyond the experience envelope of the aforementioned transplutonium production facilities [Har, 1993]. For example, the radiation resulting from the Cm isotopes in the TALSPEAK process will likely degrade the lactic acid and DTPA during the back-extraction process rendering these solvents unusable for recycle [DJM$^+$, 1993]. Further, the PUREX extractant tributyl phosphate (TBP), which has excellent extraction characteristics and availability, cannot be used because of poor radiation stability characteristics [SM, 1980]

However, damage to the reagents is function of activity, concentration, and exposure time. Although ABC conditions will be about a factor of eight that of the concept proposed for breeder reactor processing, preliminary ORGEN calculations indicate that the proposed chemical process is within the feasibility envelope, that is, varying parameters such as cooling time and concentration should allow the material limits to be met [Dew, 1993].

Given this problem, to extrapolate performance from existing experience and preliminary calculations offers a reasonable assurance that an ABC type processing system is attainable, however, this must be tempered with a note of caution. Developing the processing technology to the scale and performance required for the ABC will require considerable development time and effort. Because there is no experience at the conditions proposed for the ABC, many questions remain unanswered which will require that both cold and hot cell experiments be conducted in addition to experiments evaluating its performance as part of an integrated ABC type system. Both Bond [Bond, 1993] and Harwitz [Har, 1993] believe that the system proposed for the ABC is a feasible and worthwhile pursuit, but caution that the radiation problem posed by the short cooling times makes the concept far from being an implementable technology.

3.4.4 Secondary Coolant System

The functional requirement for any reactor cooling system is the ability to remove thermal energy generated by fissioning and convert it into usable electrical energy. In a solid-fuel power reactor, the heat generated by the fissioning of fissile atoms is removed by forced convection along the fuel rod elements, and passed onto the secondary side via a primary

heat exchanger. The primary working fluid can be one of many commonly used coolants including: light water, heavy water, carbon dioxide or liquid sodium, which, of course, are chosen depending on the specific purpose and design of the reactor. For example, liquid metal coolants are used exclusively in fast reactors where it is important to minimize thermalization of the neutrons, whereas, a heavy water coolant is preferred in thermal reactors where thermalization of the neutrons is central to the fissioning process. However, reactor concepts typically use light water for the secondary working fluid when cost and availability are the constraining parameters.

Although the ABC reactor concept varies considerably from standard reactor designs in terms of fuel form and steady-state criticality, the secondary side is essentially the same as the type employed in the heavy water moderated and cooled CANDU reactor design. In the CANDU reactor, the heavy water acts as both the primary coolant and moderator, while light water is used as the secondary fluid to drive the steam turbines [Nero, 1979].

Plant Efficiency

Because the function of the secondary side is to produce electrical energy, thermal and plant efficiencies are used as relative performance metrics for various power systems and ultimately determine the cost of electrical production. In general, the thermal efficiency of the steam power cycle is defined as the ratio of the gross electrical power output and the heat energy generated from the fission process, while the net plant efficiency is a ratio of the net electrical power to the grid to thermal energy production.

For all steam power cycles, the thermodynamic efficiency is a function of the temperature at which heat is added to the working fluid (coolant) and the temperature at which at which heat is rejected to the environment. In the case of a simple reversible carnot cycle, for example, the thermal efficiency can be written as:

$$\eta = 1 - \frac{T_r}{T_a} \tag{3.15}$$

where T_a is the temperature at which heat is added to the working fluid and T_r is the temperature at which heat is rejected by the fluid. While no power cycle is completely reversible, the efficiency of these systems remain a function of the ratio of these average temperatures. Thus, since the ambient temperature is arbitrary to the location of the

plant and time of year, a primary goal for reactor design concepts is to increase the in-core temperature as much as physically possible. For solid-fuel reactors, this temperature is limited by the structural limits of the fuel cladding and fuel. Current reactor concepts, for example, do not unusually exceed 310^{o}C, which is the average primary output temperature for pressurized water reactors [Winterton, 1981]. Fluid fuel systems are not restricted by this structural limitation.

The net plant efficiency is defined as the electric power capacity available to the grid divided by the heat production rate of the reactor, and is a function of the thermodynamic efficiency *and* the power consumed by primary pumps and other ancillary plant equipment [Winterton, 1981]. One inherent disadvantage of the ABC subcritical concept is that a significant portion of the electrical energy generated, approximately 20%, is required to power the linear accelerator. Therefore, assuming an equivalent thermal efficiency, an ABC type system cannot match the net efficiency of critical reactors. However, as a subcritical system approaches critical operating conditions, the source requirement is minimized, and efficiency difference between critical and subcritical systems approaches zero. Therefore, there is a strong incentive to design the ABC system to operate as close as possible to critical condition (K_{eff}=1), and at a high temperature.

ABC Secondary System Concept

As noted above, the ABC secondary side is simply a modified CANDU reactor cooling system [CISK, 1992] that circulates a liquid fuel, as opposed to a coolant, through the primary heat exchanger. Figure 3-18 shows the basic configuration with possible steady-state operating conditions. The D_2O slurry is circulated through the primary heat exchanger to transfer its thermal energy to an intermediate cooling loop that isolates the highly radioactive primary slurry from secondary cycle. The proposed heat exchanger geometry is a counter-flow, shell-tube with the slurry flow on the tube side. The secondary coolant is light water which is similar to any steam power generating facility. The steam, created in the steam generators, drives the turbines, condenses, and recirculates back to the steam steam generators to complete the cycle.

Commercial development of heavy water moderated reactors has been fairly widespread. The most common type, the Canadian deuterium-uranium (CANDU) reactor, employs a pressure tube type reactor core [Nero, 1979]. In this concept, the primary coolant is

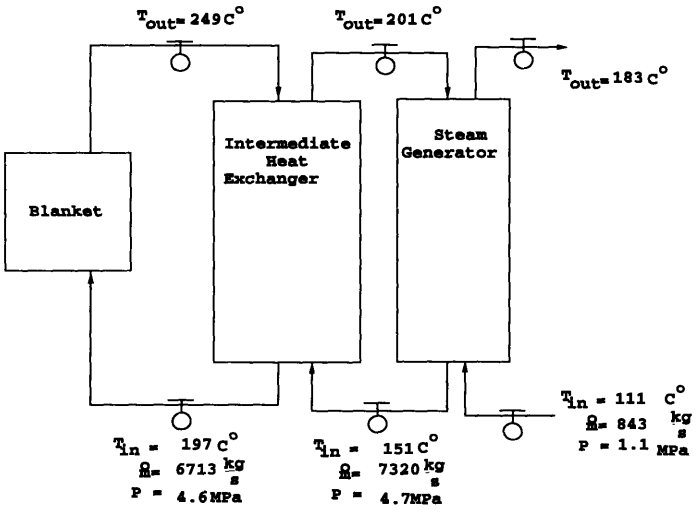

Figure 3-18: ABC secondary system with example operating conditions

circulated past the fuel in pressure tubes which allows the coolant loop to be separated from the heavy water moderator. The heavy water blanket surrounds the fuel elements and is contained in a low pressure tank. Variation on the Canadian system have been developed that use other primary coolants including, light water, organic liquid, and carbon dioxide. In a typical CANDU reactor, the heavy water coolant is maintained at a pressure of about 1500psi(10Mpa) and in passing through the pressure tubes, reaches a temperature of 590 °F (310°C). The flow rate for the 600 MWe CANDU is about 7.6 Mg/s [Nero, 1979].

Technical Maturity

Given the information above, the ABC secondary side can be said to be technically mature. Both in terms of concept and operating conditions, the ABC system is within the experience envelope of the CANDU system.

However, two points of uncertainty should be kept in mind, given that the design of the primary heat exchanger is yet undefined for systems that circulate a high temperature, high activity slurry working fluid. The first point centers around the slurry fuel and its corrosion/erosion characteristics. As discussed in greater detail in the slurry concept section, erosion is a process by which the shear force generated by the working fluid continually exposes metal to a corrosive environment. With the added abrasiveness of the suspended actinide particulate, this process will be accelerated. This effect is further augmented by the

requirement that the slurry system must be pumped through the system at high flow rates [DS, 1963]. Failure of the heat exchanger tubes could result. In fact, it is concern for the possibility of heat exchanger failure that has lead to the intermediate coolant loop. A second area of concern is the tendency for the plutonium to plate out on walls of the heat exchanger tubes. The slurry must pass through the heat exchanger that has a characteristically high surface/volume ratio. Experience has shown the importance of guarding against plating-out the oxides or the formation of cakes, which may significantly reduce the heat transfer properties of the heat exchanger.

3.4.5 Homogeneous Slurry Fuel

Homogeneous liquid reactor fuels are of two general types. A solution-based fuel consists of a working fluid containing the fissile material in solution as a salt. The slurry-based fuel consists of a two-phase mixture of heavy or light water and the fissile material in an insoluble oxide form, such as uranium-oxide, thorium-oxide, or plutonium-oxide. Ideally, reactivity control concerns require that the liquid fuel be homogeneous, however, slurry systems, by nature of its dual phase, have been know to have difficulty in this regard. This discussion centers on the slurry-type system which is the basis of the ABC fuel concept, and considers it in light of experience gained in the early 1960s, when research related to these types of nuclear fuels was conducted.

Initial interest in liquid fuel systems was sparked for several reasons. First, liquid fuels could be recycled on-line. Unlike solid fuels elements which lose their effectiveness as fission byproducts accumulate - leaving a majority of their potential nuclear energy untapped - a liquid fuel system coupled with an on-line chemical separation process could conceivably remove fission byproducts on a continual basis; allowing for a high burnup of the fissile material while reducing the required amount of fuel in the core. Researchers at the KEMA facility envisioned that continuous reprocessing would greatly minimize fuel cycle costs by eliminating the need for fuel fabrication and reprocessing [KW, 1963]. Second, liquid fuel would allow higher power densities to be achieved. Since the efficiency of a power system is limited by the core temperature, using liquid fuels would allow higher temperatures to be attained and thereby increasing the efficiency of the system. Third, it was believed that because of the negative reactivity feedback caused by the expanding fluid fuel with temperature, the system would be inherently safe and not need control rods [DS, 1963].

General Slurry Systems

At equilibrium, the slurry fuel system consists of a liquid composite of actinide-oxide particles suspended in heavy water, D_2O. The heavy water acts both to "suspend" the actinides for transport and to moderate the neutrons generated by fissioning. A slurry particle consists of a closely pack group of actinide crystallites which are bound together by cohesive Van der Waals forces [DS, 1963]. Larger less dense groupings of particles often form, called flocs, which can greatly influence the rheologic and neutronic behavior of the slurry. Thus, the ability to control the size and shape of the particle will determine whether the slurry fuel can be maintained within operating limits. Although the size of particle can be controlled during formation to be within the limits desired for slurry systems, the particle geometry is likely to change during operation [DS, 1963].

Controlling the size and shape of the particle determines how well the oxide will remain suspended in the heavy water(i.e., its stability), and therefore, affects the local actinide concentration and reactivity within the liquid. Ideally, a homogeneous slurry is desired, however, oxide settling and caking often occurs as flow velocities are not able to maintain suspension of individual particles or flocs. At a minimum, the required flow velocity must be determined so that settling in horizontal pipes does not occur [DS, 1963], although settling in stagnant flow areas are equally important to consider. Since flocs are in large part a function of particle geometry, oxide formation and control becomes a central concern in slurry systems.

Pumping at high velocities can ensure that slurry settling is minimized, however, this would also increase the rate at which erosion and corrosion takes place in the system [DS, 1963]. In fact, both velocity and particle size affect this rate. Large particles are more likely to break through the laminar boundary layer near the interface between the system structure and the working fluid and erode the oxide film on the containment metal, increasing the rate of erosion. Further, flow velocity determines the rate at which this erosion mechanism occurs for given particle sizes.

ABC Slurry Fuel Concept

Early slurry reactor concepts used the core geometry as the mechanism for achieving critical condition. A sub-critical slurry of defined concentration would flow into a chamber that,

because of its geometry, would cause the slurry in the chamber to become critical. The actinides particles would undergo fissioning producing higher actinides, fission products, and energy. The energy would be removed by circulating the slurry through a heat exchanger.

Dawson and Soden [DS, 1963] note that based on the experience at Oak Ridge, KEMA, and other facilities, the technical requirements for slurry fuel systems are extremely complex and require a balance between several critical parameters. The flow rate must be high enough to maintain oxide suspension without approaching velocities that would cause extensive erosion/corrosion in the piping. Particle geometry affects how well the oxide will remain in suspension, but also affects the efficiency by which fission products separate from the oxide particle upon fissioning. Fission products that do exit the host oxide particle bombard other slurry particles and surrounding system structure, changing their character, and therefore the overall behavior of the suspension system. In addition, the ejected fission product also causes radiolytic decomposition of the moderator material which results in the generation of volatile gases.

The ABC developers believe that such complexities can be controlled and the benefits of liquid fuel reactors can be successfully applied to the problem of weapons-grade plutonium processing. The ABC concept centers around its ability to separate and reprocess plutonium and other transmutable fission byproducts on-line. On-line processing allows the plutonium to be fissioned far beyond solid fuel system which, in most cases, would require separate reprocessing and fuel fabrication to achieve similar burnup times. In general, as the time spent in reactor increases the more plutonium isotopic conversion and fissioning takes place. Therefore, the slurry system is an integral part of the ABC strategy.

Figure 3-19 illustrates the basic layout of the slurry system. At equilibrium, the slurry consists of an aqueous mixture of plutonium, americium, curium and fission products that are suspended in a heavy water (D_2O) matrix. The D_2O serves two purposes, (1) it functions as a carrier for the actinide particles, and (2) it serves to moderate the fission generated neutrons (slows the fast neutrons to useful energy levels).

While in the blanket region, the slurry undergoes extensive fissioning due to a high neutron flux generated by the combination of the spallation neutron source and multiplication due to fissioning. The thermal energy is removed by circulating the slurry in a primary heat exchanger. Average inlet and outlet temperatures of 273°C and 325°C are being discussed [Cowell, 1994], however, these values are only for concept definition and could vary

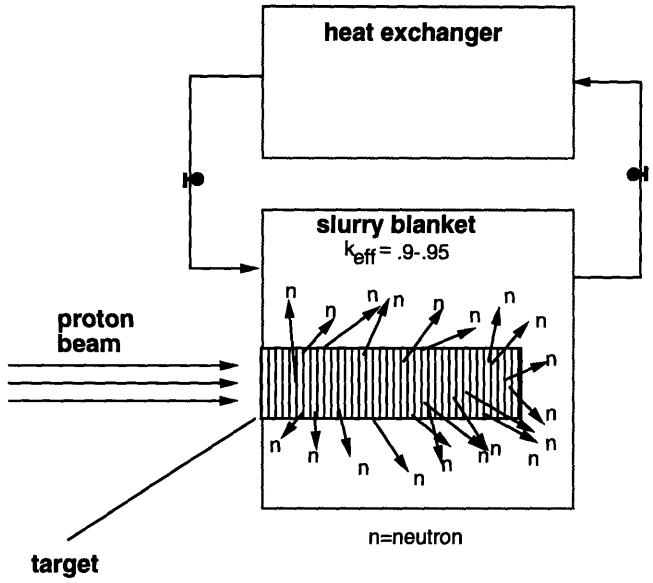

Figure 3-19: Slurry fuel circuit

considerably as development continues.

In terms of reactor design, it is important to note that for a given sub-critical geometry, the neutron flux is a direct function of K_{eff}, thus, as K_{eff} approaches a value of 1.0 (critical condition), the power generated per volume increases. Because the thermal efficiency increases as the power density, there is economic incentive to design a system as near to $K_{eff} = 1.0$ as possible. This goal is in direct conflict with the concerns to maintain subcritical conditions, thus severely limiting the ABC effort [Lin, 1993, And, 1993].

Technology Base

Much of what is known about homogeneous liquid fuels was gained in the late 1950s through early 1960s when liquid fueled reactor concepts were considered alongside solid fuel systems for various applications, such as fuel breeding and energy production. Fuel breeding concepts included a slurry matrix that would be formed by an oxide fertile material, such as UO_2, suspended in water. The slurry would be circulated into a neutron rich environment, allowing neutrons to be captured to produce fissile atoms. In a similar way, power producing systems would circulate a suspension of fissile oxides that would be brought to criticality, and then be passed through a heat exchanger to remove the energy produced from fissioning. The benefits offered by liquid nuclear fuels were thought sufficient to warrant extensive investigation in the hope that they could be developed to full scale facilities. In the United

States, at the Oak Ridge National Laboratory, both solution-based and slurry-based systems were studied, however, much of their effort was geared toward a uranium solutions fuel in which the fissile material is dissolved in the primary fluid as a salt. The most directly applicable experience in slurry nuclear fuels is found at the KEMA center in the Netherlands, were a uranium-oxide slurry concepts was studied and tested for application to power production.

ORNL

In the United States, interest in homogeneous aqueous reactors was centered around fissile fuel breeding and power production. Government-funded research dominated with several large scale experiments designed to test fuel properties and develop research scale reactors. It was hoped that these experiments would serve as the foundation for full-scale development of homogeneous aqueous reactors, theoretically promising many potential advantages. Two primary experiments were conducted: Homogeneous Reactor Experiment One and Two (HRE-1 and HRE-2). The first served as the low-power precursor to the second. Successful demonstration of HRE-1 at a power level of 1 MW prompted the second experiment in which a 5 MW power level was attained in 1955 [DS, 1963]. In both cases, a uranium solution,D_2O moderated, was used as the aqueous fuel form.

Although suspension slurry systems were also of interest during this research effort, no experiments was conducted that directly tested slurry fuels in operation. Study centered on rheological testing,i.e., flow properties and stability of oxides in suspension due to flow parameters. Also of interest is the corrosion and erosion testing conducted. Thoria slurries formed the basis of this testing to breed U-233 [DS, 1963].

KEMA

Other research, which focused directly on slurry nuclear fuels, was conducted during the same period as the Oak Ridge experiments at the KEMA Laboratories in Arnmen, the Netherlands. In their work, which was based in part on the experience gained from the Oak Ridge experiments, studied UO_2 suspension fuels. A subcritical tests reactor test was operated at 250kW, followed by a 1MW reactor test. Their goal was to determine the feasibility of using a suspension reactor with uranium dioxide as the fissile material.

The Oak Ridge HRE experience prompted the KEMA team to focus extensively on defining and controlling the geometry of the slurry particles because of the extensive interaction between particle size on slurry stability and erosion-corrosion [DS, 1963]. The

particles must be small enough to allow the fission products created by a fission event to be ejected from the particle, but large and smooth enough to minimize re-absorption of the fission products back into the solid particle phase [DS, 1963]. Also, the particles must be smooth and preferably spherical to reduce attrition and erosion during circulation [KW, 1963].

Reactivity Control

As in any fission system, controlling the reactivity of the ABC is of paramount concern, and for this discussion, is defined as a primary functional requirement. However, the slurry-fuel based system presents an entirely different controls issue than previously experience with solid fueled reactors. By nature of its design, the ABC system does not achieve a critical state, therefore, there is an excess amount of negative reactivity present in the system, that is, K_{eff} is less than unity. Since the system is not designed to operate at critical, the primary goal of reactivity control is to assure that all operations within the ABC system are subcritical, with a clear understanding of all possible reactivity behavior in the system.

As discussed earlier in this chapter, the production rate of neutrons, and therefore the power level, is dependent on several reactor system parameters. Some of the parameters such as amount of fissile material present and moderating efficiency will contribute to the increase in neutron population, while parasitic materials within the fuel and in the structure of the system itself tend to reduce the population.

Reactivity is a term that describes the state of criticality of a reactor relative to critical state. Reactivity is a way of relating the effect of changes in various reactor conditions on $Keff$. In general, reactivity is defined as the fractional change in K_{eff} and can be expressed in the following form:

$$\rho = \frac{\Delta K_{eff}}{K_{eff}} \tag{3.16}$$

.

As various parameters change, controllable or not, the rate of production and loss of neutrons, and, therefore, the condition of criticality in the reactor changes. Thus, for the ABC, the net reactivity is made up of several reactor parameters:

$$\rho_{net} = \rho_{fuel} + \rho_{poisons} + \rho_{mod-temp} \tag{3.17}$$

Where fuel reactivity is the percent change in K_{eff} of the system to changes in fuel concentration, poison reactivity is defined as the percent change in K_{eff} given a change in concentration and species of neutron poison in the slurry, moderator-temperature reactivity is the percent change in K_{eff} due to a change in slurry temperature.

Plutonium Concentration

Most of what is known about fissile-bearing fluid, and how to control their reactivity, is found in the reprocessing industry. By its nature, a liquid-fuel system, as is the type proposed for the ABC system, is more difficult to define and control than its solid-fuel counterpart. Controlling the reactivity of liquid fuel systems is not intuitive and is a function of several parameters including the concentration of the fissile material, the moderating effect of the water, the temperature of the fuel, the concentration of reactor poisons, and the geometry of the system and the density of the fluid.

In general, as the density of the fissile material decreases, the critical mass increases, therefore, it would seem that a decrease in concentration of the plutonium oxide in heavy water reduce the net reactivity. This may not be the case for a liquid-fuel system. Knief [Knief, 1985] provides an example to illustrate. For an assumed spherical geometry, critical mass of alpha-phase plutonium with a density of 19.6 $\frac{g}{cm^3}$ metal is 9.8 kg, which corresponds to a critical radius of 5.0 cm. As the plutonium is diluted in water, the density of the plutonium decreases which causes the neutron leak rate to increase, and as expected, the critical mass and radius increases. However, as dilution continues, the moderating effect of the water begins to add positive reactivity to the system. There is a point in this process, at 25g/l, that the "optimum moderating" condition is reached and the bare critical mass is only 0.9 kg!

For the ABC system, the fuel reactivity is controlled by chemically separating and recycling the plutonium resident in the slurry, and by injecting fresh plutonium oxide. However, the primary concern in controlling the fuel reactivity centers on the ability to maintain a homogeneous slurry mixture. Because of the dual-phase nature of the slurry, there is a possibility that the slurry will "settle" in low velocity regions in the loop, or plate onto the walls of the system. In fact, several types of solids formed in the UO_2 and ThO_2-UO_2 slurry circuits of the Oak Ridge HRP experiments [Cowell, 1994, DS, 1963]. They included: (1) soft plugs in pressurizer and bypass lines, (2) a thick, hard film in regions of high velocity or high impact, (3) thick hard deposits in interstices of the pump impeller,

and, (4) a thick, hard layer plating uniformly distributed throughout the system.

The implications of this behavior on fuel reactivity control are serious [And, 1993]. In the KEMA subcritical experiment [KW, 1963], the "effective concentration", which includes the settling out of particles, varied with flow rate. This behavior was caused by flocs forming in low velocity regions of the circuit, and once formed, would tend to settle, thus leaving a lower concentration of oxide in the slurry.

A second issue pointed out in the KEMA subcritical experiment was the effect of stopping the flow in the primary circuit completely. Experimenters noted that if the flow was stopped for long enough to allow the oxide particles to completely settle from the liquid, there was a danger that, if the flow was resumed, more than a critical amount fissile material would be injected into the reactor vessel.

However, although reactivity control issues are serious, the KEMA experiments seem to indicate that a suspension system can be operated, given sufficient control of the factors that cause settling and plating. For example, by carefully controlling the oxide particle geometry and, thereby, the formation of flocs, particles were easily dispersed with a low stream velocity [DS, 1963]. Another possibility is to introduce dispersing agent into the slurry [DS, 1963]. The dispersant would break up and scatter the oxide and cause it to be more evenly distributed throughout a heavy water medium. Some work on dispersing agents for high temperature systems was conducted at ORNL [DS, 1963]. Also, the system could be designed with high flow velocities to ensure that oxides particles and flocs would remain suspended. In addition, the KEMA research determined that a major factor in the formation of flocs is the pH of the system which is a controllable parameter [KW, 1963]

Slurry Temperature

The effect of temperature on the ABC slurry reactivity is an important part of the systems reactivity control process. In uranium-fueled systems, a temperature increase adds negative reactivity by broadening the resonance peaks of U^{238} (Doppler broadening). Thus, uranium provides an inherent safety mechanism that prevents a run-away chain event. In the past, one of the main concerns with an all-plutonium fuel is that U^{238}, which provides this feedback mechanism, is not present. However, for the ABC slurry, two complimentary reactivity mechanisms, both based on density changes in the slurry, provide this safety feature. First, as the temperature increases, the moderator density decreases, which causes the neutron leakage probability to increase. Second, as the density decreases, the concentration

of plutonium also decreases, which adds negative reactivity. The only existing data on the effectiveness of this feedback mechanism is found in the KEMA experiments. However, since an uranium oxide was used as the fuel, the temperature reactivity effects, which were found to be "completely effective" [KW, 1963], were a combination of the Doppler and density effects noted above.

Another area of uncertainty with regard to temperature reactivity effects of a slurry centers on the "magnitude" of the reactivity change. Although the addition of negative reactivity is desirable to counter increases in power, in the case of power decreases, an equally positive reactivity insertion will occur. There is concern that, for a uranium slurry, going from hot operation to cold shut down may insert positive reactivity beyond what the system is able to control [MM, 1993]. This concern was also expressed by researchers at the KEMA subcritical experiment who stressed the need to reduce the variation in temperature of the slurry [KW, 1963] However, since the ABC system will have a smaller temperature reactivity effect than a uranium based system, this may not be a concern.

Fission Products

A fundamental design feature of the ABC system is that fission byproducts that build up in the slurry will be continually removed as a means of maintaining the desired K_{eff}. With the large neutron flux that is expected, the generation rate of the fission products is likely to be much larger than in current, lower flux reactors [Rabotnov, 1992]. There is some concern that the higher generation rates, especially for the Xenon and Samarium-149 fission products, will pose reactivity control problems for the ABC. For current LWRs, the effect of transient Xenon and Samarium concentrations are countered by controllable excess positive reactivity such as control rods. Since the ABC does not have these controls, and must rely solely on waste removal as a means of reactivity control, problems may result [MM, 1993].

Samarium is produced in the Nd decay chain. Nd, with a half-life of 1.7 hours, decays to Pm. Then, with a half-life of 53 hours, Pm decays to Sm-149. The Sm-149, which has a large absorption cross section, absorbs a neutron and is converted to stable Sm-150. Thus, the level of Sm-149 is determined by both the Pm decay rate and the neutron capture rate. In a standard reactor, an equilibrium concentration is reached in about 40-50 hours [NCSU, 1990]. However, when the reactor is shut down, the neutron capture mechanism becomes zero while the Pm-149 decay rate remains constant, thus the level of Sm-149

concentration continues to rise until a peak value is reached. The subsequent reactor start-up will have to compensate for this negative reactivity buildup. The primary concern with this fission product is that, given its large absorptions cross section and large generation rate in the high flux system, it may be difficult or impossible to "restart" the ABC system [Rabotnov, 1992, MM, 1993].

Xenon

In a fissioning reactor, Xenon is created in two ways: the decay of iodine-135, and directly as a fission product. Since Xenon decays rapidly and by neutron capture, its effects are temporary, but extreme, and its corresponding negative reactivity has a large impact on the net reactivity of the system. Because of the expected power densities of the ABC system, this is of particular concern [MM, 1993].

To understand, consider a system in which the source and decay terms for the Xenon are balanced and the concentration in the core is in equilibrium. Now, assume that a step power increase is introduced. Initially, the increase in power results in a decrease in concentration as the xenon is consumed, and reaches a minimum value in about 2 to 6 hours. However, after some time, the iodine concentration, from which Xenon is created, builds up to a new higher level corresponding to the new power level. Because the concentration of iodine is larger than the initial amount, in about 40 hours, the Xenon concentration reaches a new higher equilibrium level which corresponds to the higher power level.

A power decrease has a similar but opposite effect on the xenon concentration. Consider a system that suddenly shuts down, after being at some power level. Initially, the burnup term of the xenon becomes zero which causes the level of the xenon to rise, i.e. there is still xenon being produced from iodine decay. The concentration of Xenon continues to increase until about 2-6 hours, when the effect of a diminishing concentration of iodine - no longer being generated by the reactor - begins to show. Eventually, about 70 hours after shutdown, the xenon concentration is diminished. The value of xenon reactivity, at its peak after shutdown, is about twice the equilibrium value at power [NCSU, 1990].

Woosely *et al.* conducted a series of reactivity calculations to estimate the dynamic behavior of an actinide slurry for an ATW system, in an effort to estimate the possibility of unstable positive reactivity growth due to actinide composition changes, temperature changes, and changes in Xenon concentration. They conclude that the two prominent reactivity effects due to changes in reactor power is the Xenon oscillation and temperature

reactivity effects. They conclude that because the maximum increase in reactivity due to power increases was .01, the system "could require active measures for compensation." For the Xenon calculation, they assumed a K_{eff} of .95.

However, Woosley indicates that in case of large additions of negative reactivity, i.e., increases in concentration of Samarium, or Xenon, the ATW (or ABC) system, by nature of its subcritical operation, does not require a delay in start-up while the xenon decays or Samarium is cleaned from the system. The decrease in negative reactivity due to these fission product will result in a lower K_{eff}, which means that, for a given source input, the system will operate at a lower power level than prior to the shut down, but not prevent start-up.

Other Issues

Other concerns, not directly related to reactivity control of the slurry, also have the potential of impacting the performance of the slurry system, and, therefore, need to be mentioned. The first issue concerns the corrosive and abrasive nature of the slurry, in particular the oxide particles, which could, over time, cause catastrophic failure of the primary loop containment. The second issue, appropriately, is emergency cooling and containment of the primary slurry system. Because the fuel is in liquid form and is circulated at high pressure and temperature, extensive containment and emergency cooling schemes will have to be employed to ensure that the risk to the worker and the public is not compromised. Finally, the third area of concern is the generation of explosive gases by radiolytic decomposition of the heavy water, which will require that some method of removal or recombination be employed.

A significant concern with using a slurry fuel is its highly corrosion nature, which, when coupled the high velocity particle stream, erodes the metal surfaces of the primary loop. This destructive process is carried out by two mechanisms: (1) a chemical attack on the metal surface by oxidizing the metal which then forms an metal oxide layer between the metal surface an the slurry, and (2) an abrasive attack on the oxide layer by the slurry particles which re-exposes the metal surface for further oxidation. Left unchecked, this process would wear through the metal containment and spew radioactive material into secondary containment. The main factors that affect this process include [DS, 1963]:

- concentration - as the concentration of oxide particles increase in the system, the rate of erosion increases,

- temperature - temperature promotes the oxidation of the metal, and
- flow velocity - ORNL experiments verified that an increase in flow velocity increases the rate of attack, erosion, of the metal.

In addition to these factors, Cowell [Cowell, 1994] states that a dominant variable to this process is particle size. As the particle size increases, the abrasive nature of the slurry increases.

As noted earlier, the most recent data on slurry erosion/corrosive behavior comes from the KEMA tests. The KEMA researchers, considering the results of the ORNL experiments, concentrated their efforts in controlling the oxide particle geometry as a means of controlling the slurry stability and corrosive/erosive behavior of the slurry. As a result of their efforts, the erosive character of the slurry was reduced as compared to the ORNL experiments[Cowell, 1994]. The particles for the KEMA experiment were prepared using a sol-gel technique which produced roughly spherical shaped particles, which in turn made the particles less abrasive. However, erosion remains to be a problem in sharp bends in piping and orifices.

Safely containing a fission-product laden slurry in the event of primary containment failure, is a significant concern, even more so than for standard, solid-fueled reactors [MM, 1993]. In response to the work done at Oak Ridge and KEMA, Dawson and Sowen [DS, 1963] conclude that because the fission products are flowing through the entire primary loop, unlike a solid fuel reactor which contains its fission products in the fuel pins, the entire primary slurry system will require a "very high standard of containment throughout the whole circuit." This conclusion was echoed by KEMA researchers who recognized that if a breach in the system were to occur, the slurry would "spread readily from the system." Therefore, they concluded that "very high standards of the leaktightness" of the primary system must be guaranteed [WH, 1971].

In the event that a loss-of-fuel accident were to occur, the ABC concept proposes that the uncontrolled release of primary fluid could be reduced by draining the fluid from the primary system. However, this process must be carried out faster than the loss of pressurized fuel. Moreover, the slurry that is successfully drained from the system will contain highly active fission products which will require extensive cooling. Therefore, safely containing a loss-of-fuel accident remains a serious outstanding concern for the ABC design and can only

be resolved as part of a detailed development program.

Radiolytic Decomposition

Radiolytic decomposition refers to the degradation of the moderator molecule by a high energy fission fragment. In this reaction, the energy from the fission fragment breaks the water molecule into its constituents, and thereby producing D_2 and O_2 gas. In the ABC system, this reaction is critical because the fission fragments come in direct contact with the heavy water, by design, whereas, solid fuel reactors maintain the fission fragments within the fuel element. Since this process is proportional to the reactor flux, the generating rate of hydrogen gas is expected to be relatively high for an ABC-like system. Cowell [Cowell, 1994] notes that for a thorium breeder design, a variation of the subcritical design, the decomposition rate is expected to reach 10 moles of hydrogen per hour per liter of slurry.

The volatile and radioactive character of these gases makes their removal from the system extremely important. In addition to being highly radioactive due to the tritium component [Rabotnov, 1992], the gas is extremely volatile and prone to chemical explosions [MM, 1993]. Also, high concentrations of radiolytic O_2 and D_2 could potentially increase the rate of oxidation of the systems metal surfaces [MM, 1993].

One way to reduce these effects is to remove the fission products that have been ejected from the actinide particles. The KEMA team proposed a concept in which a "scavenger" material would to be injected into the slurry that would then absorb the fission products. In this way, the fission products could be removed from the slurry [DS, 1963]. In an effort to recombine the gases, Cowell[Cowell, 1994] note that there are two possible options to accomplish this: (1) remove the gas from the slurry, recombine, and inject back into the system, and (2) add a recombination catalyst directly into the slurry which would reduce the accumulated levels of the gases. In the KEMA experiment, an external recombining system was employed, although it experienced various problems [Cowell, 1994].

3.5 ABC System Summary

In the previous sections, the details of each ABC system component is given, including a description of basic principles and technology base. It was shown that in varying degree, these components rest on proven concepts, i.e., the "physics" of the technology is defined. However, these technologies have not been developed to the capacity and for the environ-

DP#	Hardware (DP)	Function (FR)	Technical Base	Notes:
111	Linear Accelerator	Provide proton source.	Demonstrated capacity on limited scale.	Accelerator technology proven for research applications. Production systems not demonstrated.
112	Spallation Target	Provide spallation neutrons.	Demonstrated capacity on limited scale.	Target concept proven for research applications. Power density is main technical concern. No significant technical issues.
113	Oxide Slurry Fuel	Provide neutron multiplication.	Demonstrated capacity on limited scale.	Limited technology base. Operating history shows several technical concerns. Extensive development work necessary.
121-2	Modified CANU Secondary System	Remove thermal energy and convert to electrical energy.	Demonstrated full capacity, mature technology.	Fully demonstrated system with no significant technical concerns in ABC concept.
131	Solvent Extraction	Isolate actinides for recycle.	Demonstrated capacity on limited scale.	Limit scale production history. There is concern that the scale and extreme conditions in the ABC present significant problems.

Table 3.3: Technology Maturity of Components

ment needed for their role within an ABC system. Integrating these technologies into a system that transmutes actinides constitutes an entirely new and innovative application.

This section provides an overview of the ABC concept, describing how each major components affects the functional requirements defined for the ABC system. This in turn will give an indication of what technical issues are most critical for the ABC, what conclusion can be drawn as to the system's technical maturity, and how these questions are linked to the overall goals defined for plutonium processing. Although a one-to-one correspondence between hardware component and function is desired [Suh, 1990], the ABC system, as with most large-scale systems, are highly coupled in their function, making both the design and evaluation difficult. Figure 3-20 illustrates the connection between the ABC system requirements and the primary components that make up the ABC concept, and provides a direct way to see the link between the ABC hardware and the requirements they are defined to meet. Table 3.3 lists the subsystems and summarizes the conclusions reached with regard to technical readiness for the ABC system. The details are discussed below.

Proton Source. A proton source of sufficient intensity is the first necessary ingredient in producing the high intensity neutron source that forms the basis for the ABC sub-critical

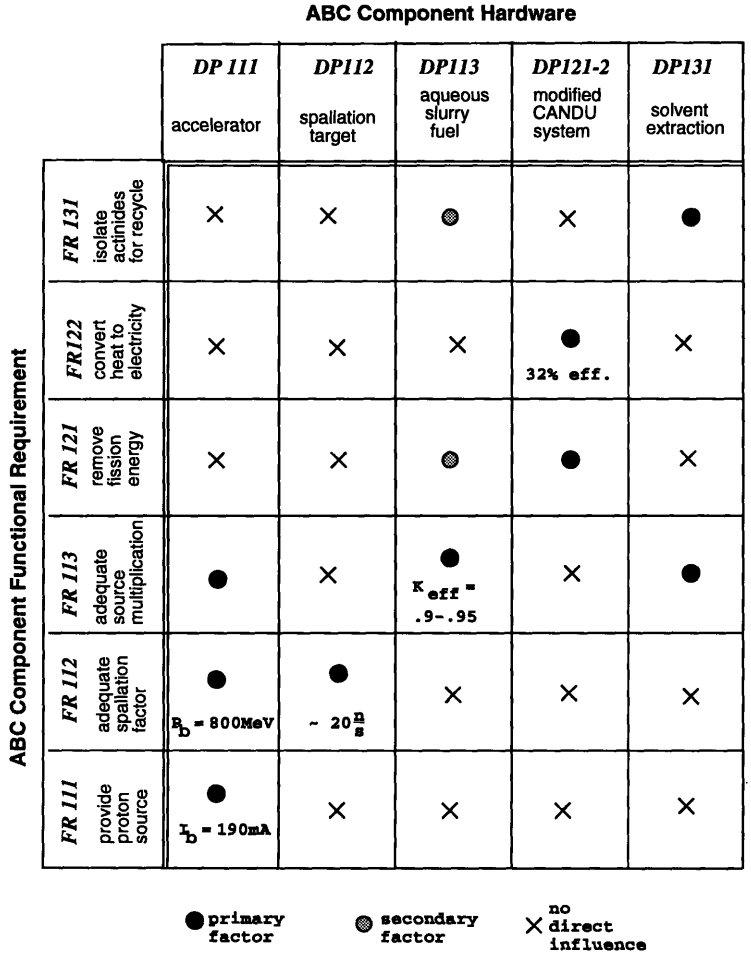

Figure 3-20: ABC system requirements and corresponding hardware

system. Source protons initiate the spallation process by colliding into a heavy metal target from which numerous high energy neutrons are generated. The amount of neutrons available define the power of the sub-critical system. The only component that directly affects this requirement is the linear accelerator, which provides a continuous stream of protons to the target. The rate of protons into the target is simply the current of the accelerator, that, for the ABC concept, is 190mA at an energy level of 800 MeV. To date, no system has achieved continuous operation at the current and energy levels required, although there is extensive experience with systems as lower currents in pulsed operation. Therefore, the accelerator component will require significant development work.

Spallation Factor. The spallation factor is a measure of how many neutrons are produced when a high energy incident proton collides into a heavy metal target. Two ABC components influence the spallation factor. First, the linear accelerator directly affects this requirement because the spallation yield is a function of the proton energy, i.e., the beam energy. The yield for most heavy metals varies linearly with the energy level of the incident proton. Second, the spallation factor is a function of the material and geometry of the target. The target design must be optimized to maximize the yield, while considering the energy density resulting from the high intensity (high current) proton beam. Because the need for the high current, proton beam specifications must be reconciled with the power density limitations of the target. The present design goal is to achieve approximately 18 neutrons per proton. Early experimental work confirms that this goal is reasonable. The main concern for this component is the power densities that are likely to be achieved with the proton beam. Verification of this component can only result from a pilot facility that is capable of delivering protons beams comparable to an ABC system.

Source Multiplication. The source multiplication, or reactivity, is the most difficult and uncertain parameter of the ABC system to define or control. As discussed earlier, it is the state of reactivity in the slurry fuel that defines the overall neutron population, and therefore the power level and rate of actinides transmutation. The difficulty of reactivity control is exacerbated by the direct influence of several ABC components on the reactivity of the slurry. By design, solvent extraction process serves as the primary means of controlling reactivity in the primary loop by maintaining tolerable levels fission byproducts including higher actinides and rare earth fission products, while reforming processed actinides into oxide particles for further transmutation. In addition, slurry system conditions such as

particle size, pumping speed, pH, temperature also affect the reactivity of the system. As noted earlier, these effects are not presently well defined. Finally, the linear accelerator affects the slurry reactivity through its effect on power density, and thus the temperature. The negative reactivity added by the reduction or loss of power, and subsequent temperature drop, is a major concern in the ABC concept.

Remove fission energy - Energy conversion. Removing fission energy and energy conversion have been treated as separate requirements, but they are really the input and output of the same process, as defined by a power system's secondary side. They are discussed together here. The requirement to remove fission energy from the primary loop and convert it to electrical energy is influenced by two ABC system components. The CANDU-like secondary side, of course, is the main factor on this point. It was chosen because of a well established technology base and direct applicability to the ABC system which uses a heavy water matrix for the slurry. The slurry system also affects the cooling process. The slurry system defines the temperature of the of primary loop, which is the principal factor in thermal efficiency. In addition, the slurry affects the thermal efficiency in its tendency to plate-out actinides and fission products onto the walls of the primary heat exchanger, thus changing its heat transfer characteristics. This effect was noted by the Oak Ridge and KEMA studies. Of all the components, the cooling system is the most mature. Concept and capacity needed for the ABC system have been demonstrated.

Isolate actinides for recycle. The primary function of the ABC is the transmute plutonium. The ABC system proposes to accomplish this by continuously separating the plutonium and other transmutable actinides from highly radioactive fission byproducts and recycle them for continued fissioning. An on-line chemical reprocessing system is proposed, using well established solvent extraction technique to isolate and recycle. In addition to the chemical process parameters, the ability the separate and recycle the plutonium is influenced by the character of the slurry fuel. The connection is brought about by the high burnup achieved in the high flux system. Higher burnup of the actinide slurry translates into higher radioactivity. In a high radiation environment, reprocessing is made much more difficult because radioactivity tends to breakdown the chemical processing agents, and, therefore, longer cooling times are necessary before the slip-stream actinide slurry can be processed. This issue is particularly important in the ABC system that proposes to process a slurry fluid drawn directly from the primary loop and cooled for a minimum time. Therefore,

although the separation concept does not extend beyond presently established technology, the ABC chemical separations concept will require a substantial development effort.

Given this information, the following points can be made:

- A benefit of the ABC system is its characteristically high thermal neutron flux (10^{15} to $10^{16} \frac{n}{cm^3 s}$). The high thermal flux allows the system to operate with a smaller inventory than is possible with current reactor concepts. The high-flux, low-inventory design means that the system operates at a high power density. Since the fuel is an aqueous slurry, the temperature constraint placed on solid fuel systems are not applicable.

- A benefit of the ABC system is that it operates at full power in a subcritical state. Because neutrons for steady-state operation are supplied from a source independent of the fissioning process, i.e., the spallation target, the system can be run subcritically to produce power. As fission byproduct poisons build in the system adding negative reactivity, positive reactivity can be added by increasing the fissile plutonium concentration. The level of sub-criticality is completely a function of how these two parameters are controlled, and a system can be designed so that source requirement is minimized, i.e., the K_{eff} approaches unity, or the K_{eff} can be reduced to add a greater sub-critical safety margin. Given this, it is conceivable that a system could be designed to run with high concentrations of fission poisons such as higher actinides and fission products, thereby transmuting them. Or, a system could be designed to have low concentration of the same. In either case, the net reactivity of the system is the sum of positive reactivity provided by the fuel and the negative reactivity provided by the reactor poisons. In all designs, K_{eff} would have to be sufficiently below unity to ensure that safe operation is maintained.

- The ABC system consists of subcomponent concepts that fundamentally sound. However, the capacity required for some subsystems within an ABC system may be difficult to achieve, and to determine their merit will require an extensive development effort. There is significant uncertainty, for instance, in whether the slurry-type concept can be sufficiently controlled to maintain safe subcritical operation. Other liquid-fuel concepts may prove to be better in this regard, however, the dual-phase aqueous slurry has the most developed separations technology base.

- Conceptually, the ABC concept is feasible, however, it represents an entirely new reactor concept with little "direct" experience from which to benefit. Therefore, the ability to design and build a production ABC system is uncertain, and will require development program that not only develops each ABC component to the maturity necessary for implementation within an ABC system, but also, in parallel, address the integration issues unique to this system.

Chapter 4

Policy Options

In the first three chapters, the problem posed by the growing plutonium stocks in Russia was discussed, a framework was proposed with which plutonium process technologies could be evaluated, and finally, a detailed look at one proposed concept, the ABC subcritical system, was conducted. With the ABC concept and associated technology base presented in detail, we now consider the ABC's proposed capabilities in light of other competing options. However, this discussion will also go beyond the weapons-plutonium problem, and consider what impact new technologies, such as the ABC system, could have on the broader issue of the total accumulation of plutonium, weapons-grade and commercial-grade.

There are a variety of options, including both developed technologies and new concepts, that could conceivably be used to process the weapons-grade plutonium, and these technologies, for the most part, are derived from concepts that are already be evaluated for other applications. Modifications are proposed that optimize each to deal directly, or indirectly, with the plutonium issue. Generally speaking, the decision will be whether to store the WGPu in pit form, their present state, for an extended period, dispose of the plutonium as waste through vitrification processing, or use the plutonium as a reactor fuel [DOE, 1993b]. In the end, it is clear that whatever option is chosen will require a substantial investment in national resources and time, in the order of billions of dollars and several years, thus, the decision process promises to be a long and difficult one, and with much debate. Ultimately, a choice will have to be formulated to balance what the economist would argue is the "the bottom line" with what is best for the international community in the long run, i.e., the reduction in risk that nuclear weapons will be used by anyone.

As many as there are concepts with which to process the WGPu, there are also proposed criteria to compare them. Evaluating a technology for plutonium disposition is difficult primarily because assumptions made to define a particular metric of comparison, for the most part, determine the outcome of that comparison. This issue does not occur in a vacuum, and decisions are being formulated with concern beyond that defined specifically for WGPu. For example, waste management is an issue. How will the investment in technologies for processing plutonium impact the effort to dispose of the large stocks of radioactive waste stored in government and commercial temporary storage facilities? Nevertheless, it is clear that the decision on what plutonium processing technology to support has to be made with a clear picture of the immediate needs specific to WGPu disposal. The "appropriateness" of a proposed concept will therefore be determined on this basis. However, the decision for one technology over another does not negate the potential of the losing concepts for other applications. Thus, the job of the policy maker is to invest in the right technology for the right application.

4.1 Decision Tree

As discussed in Chapter 2, the criteria that is likely to be used to evaluate prospective plutonium processing options are derived from three main concerns: (1) the type and extent of physical diversion barriers that are achieve by the process, (2) the unit cost to process the plutonium, and (3) the availability - or technical readiness - of the concept. Since the relative importance of these criteria have not been specifically stated by either Russia or the United States, the order presented here is not intended to indicate priority.

Several concepts to process WGPu are under consideration. In general, these concepts fall into two main categories: (1) the fission option which uses a nuclear reactor to "burn" the plutonium, and (2) the waste treatment option which mixes the WGPu with radioactive or chemical "spoilers" before permanently disposing of the mixture in the form of glass logs. Thus, the first decision point is between fission and non-fission options, as shown in Figure 4-1. From this point, each category has various individual concepts, each offering some variation on the general concept. The "form" taken by WGPu when it is permanently stored will depend on what path is eventually followed in this decision process. Thus, each concept can be judged on its merit with regard to the resulting physical form of the

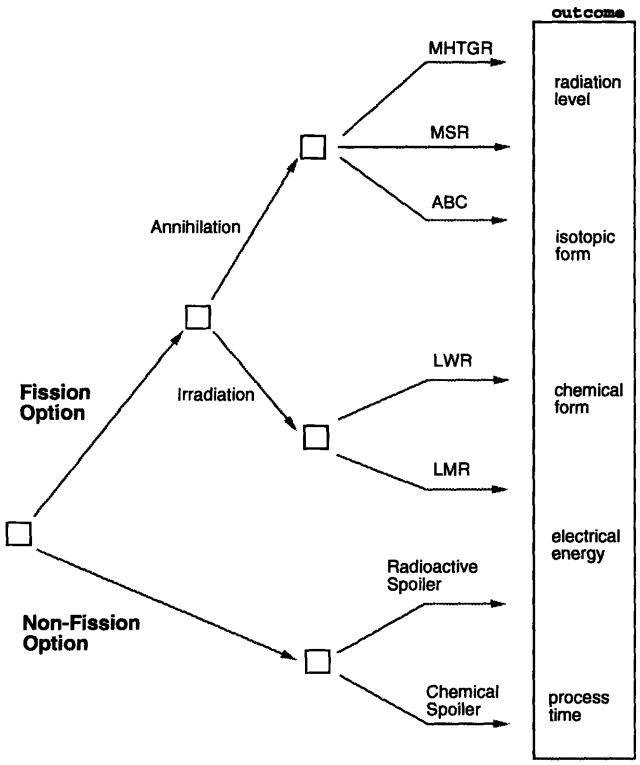

Figure 4-1: Plutonium Disposition Decision Tree

plutonium (i.e.,radiation level, isotopic character, chemical form) and the process character (electrical generation and process capacity).

4.1.1 Vitrification Options

The most promising of the non-fission plutonium processing options is to place the plutonium into glass logs for permanent disposal. Before vitrification, the plutonium is mixed with "spoilers" to render it unattractive for bomb production. The most proliferation-resistant spoiler is fission product wastes from reprocessing. The fission products are highly radioactive and would make the plutonium inaccessible except by remote handling facilities. The glass matrix would then be buried in a geologic repository.

Variations to the fission product spoiler include chemical neutron poisons, such as boron, that would render the plutonium less attractive as a bomb material, but this concept does not provide the high radiation barrier of fission products. A third option is to vitrify without adding poisons [OTA, 1993]. The least expensive of these is direct vitrification, without spoilers.

Vitrifying the plutonium is the nearest term technology considered [OTA, 1993]. However, opposition to the concept centers on the fact that the plutonium is treated as a waste without extracting its potential benefit as a fissile material for energy production. This is especially critical in Russia where plutonium-burning reactors are highly regarded.

4.1.2 Fission Options

The basic concept behind all reactors designs is the controlled release of fission energy. Variation in cooling schemes, moderating material (or use of a moderator), and types of fuel used, differentiate each reactor design. Whereas original reactor concepts used only uranium based fuels, fuel concepts have progressed to a point where a mixture of uranium and plutonium can be used. In this scheme, uranium constitutes both a fissile material (the U^{235} component) and fertile material (the U^{238} component), while plutonium is a fissile material. In the fission process, the fertile atom undergoes neutron capture, which produces new plutonium. Because the goal in using plutonium as a nuclear fuel has always been coupled with a strategy in which producing additional plutonium is desired, full core plutonium designs have not been developed to commercial scale. Therefore, the goal of burning the plutonium offers an entirely new design goal for reactors. Until now, no reactor has ever been designed specifically to minimize plutonium generation or maximize plutonium destruction.

The fission option can be thought of as having two categories of choices: (1) irradiation and disposal as a spent fuel, and (2) substantial or complete annihilation [OW, 1993] and waste disposal. In the first case, the plutonium is mixed with uranium and placed into existing reactors for fissioning. Some of the plutonium is destroyed, but a substantial amount remains. In the second case, the plutonium is placed in a reactor, but a substantial amount of the original plutonium is destroyed. This high plutonium consumption is accomplished by one or more methods. First, by increasing the plutonium residence time in the core (i.e. high burnup) by design or by recycling will increase the amount of plutonium is consumed. Second, a fuel scheme that requires only plutonium as the fissile fuel will eliminate the production of new plutonium by U^{238} neutron capture. A recent DOE study of plutonium fission reactors confirms these effects [DOE, 1993b].

Irradiation Processing

In the basic irradiation process, a light-water reactor (LWR) is partially loaded with a mixed oxide fuel (MOX), i.e., UO_2 mixed with PuO_2. Varying amounts of MOX can be loaded into an LWR, however, the most commonly considered scheme is a core containing 1/3 MOX fuel, with the remainder loaded with low enriched uranium. With this loading, power densities and control distributions will be within the operating envelope of an all-uranium fuel cycle. In general, as the amount of plutonium place in the core relative to uranium increases, i.e., higher MOX core loading, the less *new* plutonium is produced by the U^{238} neutron capture, and therefore the net plutonium destruction will increase. However, U^{238} plays an important role in the safe control of a nuclear reactor which makes the proposition of replacing it a difficult one to realize. Specifically, U^{238} offers a negative feedback effect to temperature increases in the fuel which keeps the reactor from sustaining a run-away reactivity excursion. Power increases result in an increase in fuel temperature. As the temperature increases, the probability of resonance capture by U^{238} increases, which adds negative reactivity into the system. This, in turn, counters the positive reactivity driving the power excursion. Thus, if a core is designed without U^{238}, a replacement material must be found that offers similar negative reactivity feedback. In terms of proliferation barriers, MOX can be considered comparable to spent fuel from an all-uranium fuel cycle.

The liquid metal reactor (LMR) concept has been considered as an alternative to the LWR for the irradiation process. Typically, LMRs have been designed to breed plutonium from MOX fuel by producing neutrons in the fast energy spectrum. In the breeding scenario, the rate of plutonium production from the U^{238} is greater than the fissioning rate of the feed plutonium. However, the plutonium-burning concept reverses this process by reducing the production rate to less than the fissioning rate, therefore, achieving breeding ratio of less than one. The DOE evaluated this concept and found that LMRs with an MOX fuel could substantially degrade the isotopic quality of the plutonium, i.e, increase the Pu^{240}content to about 30%; however, found that the total plutonium destroyed was limited [DOE, 1993b]. They further concluded that the ALMR concept would require development work to overcome technical and safety issues, and therefore increase the cost and schedule risks for plutonium disposition.

Annihilation Processing

The concept of accomplishing near complete burnup of the plutonium, or annihilation, comes from the idea that, by removing the breeding source of plutonium, U^{238}, and in some concepts, recycling the existing plutonium, complete burnup of the plutonium can be accomplished. However, such concepts are very new relative to those discussed above, and only in the conceptual stage. The DOE concludes that to destroy most of the plutonium in a reactor will require development of new reactor concepts [DOE, 1993b]. Extensive burnup could be accomplished with MOX schemes, but with the plutonium production also a factor, recycling, perhaps more than once, would be necessary to achieve the levels of burnup conceivable for those systems that do not generate new plutonium.

The first type of full-plutonium core reactor concept is the modular, high temperature gas reactor (MHTGR). The MHTGR is a concept that proposes to burn a full plutonium core consisting of coated plutonium oxide particles along with a beryllium oxide burnable poison. PuO_2 replaces a $U0_2$ fuel pellet that has been the baseline for concept. The small oxide particles are coated with "impervious" material and formed into a graphite matrix [Cameron, 1982]. High burnup of the plutonium fuel is possible because of the large excess reactivity from the plutonium. As the plutonium is fissioned, and fission byproducts begin to add negative reactivity, the beryllium oxide poisons can be removed allowing for longer fuel residence times. The system could be designed for various burn times, and would be a function of the excess reactivity present in the core. The MHTGR concept would use 4MT of plutonium per year for the required half-core reload [DOE, 1993b]. The spent fuel from this process would be disposed as waste. Recycling would not be needed because of the high burnup in the first cycle. The DOE plutonium disposition report notes, however, that the MHTGR concept requires development to answer substantial technical and safety issues, and this would increase cost and schedule risks [DOE, 1993b].

It is in this group of all-plutonium core reactors that the ABC and the Molten Salt Reactor (MSR) find their place. However, because these systems use a fluid fuel concept, the all-plutonium fuel can be extracted, cleaned and returned for further fissioning on a continuous basis. It is conceivable that this process would result in near complete destruction of the plutonium. The fundamental difference between the two fluid fuel concepts is that the ABC is based on an aqueous two-phase slurry fuel, while the MSR is based on solution fuel with the fissile material dissolved as a salt. Of the reactor concepts, the fluid

fuel systems are the most advanced and therefore, the least developed.

4.2 ABC Considerations

4.2.1 Diversion Barriers - vs - Technical Maturity

As proposed in Chapters 2 and 3, the primary objective of the WGPu processing is to make it resistant to diversion by terrorist or sub-national groups by creating barriers against its use in a nuclear device. The types of physical barriers include: (1) Pu^{240} or higher plutonium isotopes in high concentrations (isotopic barrier), radioactive fission products and/or chemical poisons, and substantial annihilation (extreme case of isotopic barrier).

To compare technology concepts on their relative ability to meet the first criteria, one might first attempt to determine what isotopic quality, radiation level, and/or chemical processing would render the plutonium inaccessible to such groups. Although, one could speculate on what levels are adequate, developing a technology to reduce a threat that is not clearly defined makes this approach ambiguous, and the choice of what technology to develop would be strongly dependent on the *perception* of proliferation risk [OW, 1993]. There is a another way.

Figure 4-2 shows the amount WGPu that is to be processed, approximately 150 metric tons, relative to the 800 metric tons of RGPu that rests in commercial spent fuel throughout the world. Thus, a rough estimate is that only 16% of the total plutonium, the WGPu, is being considered for disposition processing. The remainder of the plutonium, that in spent fuel, is being stored indefinitely at reactor site cooling ponds pending resolution of a permanent disposal option for commercial nuclear wastes.

As was discussed in Chapter 1, for crude bomb designs, the differences in quality between RGPu and WGPu is not significant. If the threat of diverting commercial spent fuel assemblies for bomb programs is deemed significant, then the optimal strategy would be to process the WGPu and RGPu to a level that eliminates the risks posed by both [OTA, 1993]. However, since the issue at hand is to what extent the WGPu should be processed, then the answer is straightforward: render the WGPu such that it equals the characteristics of RGPu, which according to DOE, means that the proliferator would require: (1) remote handling capability (hot cell), (2) a large scale separation facility (a function of desired processing rate), and (3) extensive capital investment to develop infrastructure, in order

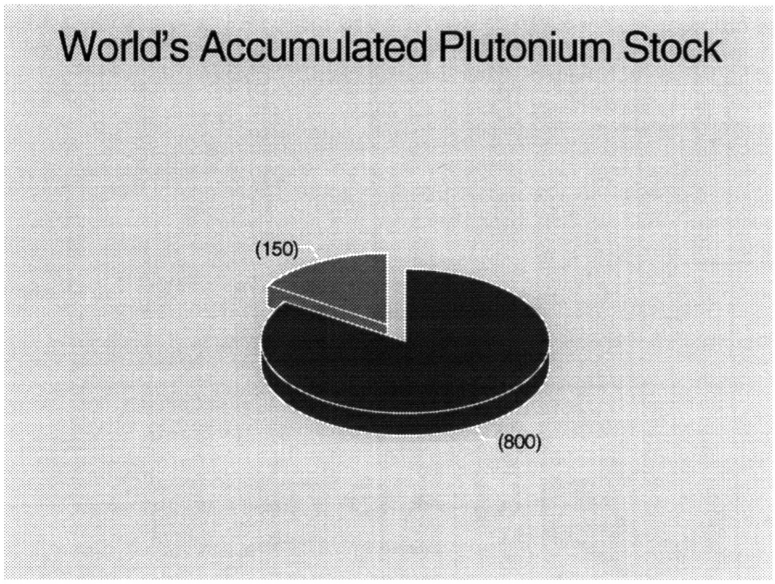

Figure 4-2: World Plutonium Stock for various technologies

to process the fuel assembly. According to the DOE report, these characteristics make the plutonium "strongly proliferation resistant" [DOE, 1993b].

Given this definition, how do the various processing concepts compare? Results published in both the OTA and DOE [OTA, 1993, DOE, 1993b] plutonium disposition studies reach a similar conclusion: a light water reactor using an MOX fuel will render the WGPu as proliferation resistance as commercial spent fuel. In addition, the OTA, which also considered non-fission options, concludes that vitrification of the plutonium with an appropriate poisons is a reasonable near-term option. Moreover, the OTA found that the least costly processing option was to mix the plutonium with glass without radioactive or chemical spoilers [OTA, 1993]; however, this option offers only minimal proliferation resistance.

As discussed above, if the ABC system is developed, it would be capable of near complete elimination of the plutonium, far beyond what is possible with MOX fuels. However, base on the assumption that the barriers found in commercial spent fuel is sufficient to prevent diversion by terrorist organization, the ABC system would not be a reasonable technology to develop when several mature technologies are capable of the task. In general, the technologies that could convert the plutonium beyond that found in commercial spent fuel would take in the order of decades to develop and would costly [OTA, 1993]. Therefore, the DOE study [DOE, 1993b] concludes:

> [W]hile the choice of more mature concepts may result in a lesser capability

for net plutonium destruction, such a choice would require little or no further development. Consequently, a technically mature concept can be expected to be less costly (relatively), and revenues from electricity production could be obtained sooner. The mismatch between cash outlays and revenues, both in time and amount, fundamentally determines the economics of any strategy for plutonium disposition.

4.2.2 Energy Production

The final point of comparison between options is the requirement that the disposition process includes the ability to generating electrical power from the plutonium. It is estimated that 100 MT of WGPu is equivalent to a three-year supply of fuel for all currently operating U.S. nuclear power generators (assuming annual refueling of one third of each rector with MOX fuel and disposal as spent fuel of whatever material is not burned) [DOE, 1993b]. To many in Russia, and even some in the US, burning the plutonium in a system that produces electrical energy is a foregone conclusion. With this in mind, the ABC, as an advanced reactor concept, is a reasonable option. However, as in the case of the proliferation barrier requirement, other, more mature, technologies could accomplish this requirement in a shorter time and lower development costs. Although the potential benefits in terms of waste reduction and subcritical operation may be attractive features to pursue, the incentive to develop the ABC concept will have to be more than for an alternative power system.

4.3 World Plutonium Inventory

As a technology for solving the specific, near-term problem of WGPu disposal, the ABC system was found to be innovative, but with an insufficient technical base to compete with the mature reactor and vitrification concepts. The argument was made that since a large percentage of the plutonium existing in the world is found in the form of RGPu, in commercial spent fuel, to convert the WGPu to a form that is more resistant than the commercial spent fuel would not make sense. Indeed, this is true if the assumption is made that spent fuel is sufficiently proliferation resistant. However, noting that a few kilograms of RGPu can be formed into a crude nuclear device [Mark, 1990], consider a case where a strategy for controlling the entire accumulated plutonium stock in the world, both weapons and reactor grade, was necessary. Within this problem definition, what technology

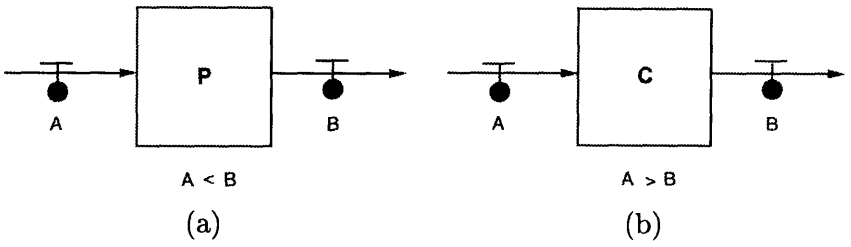

Figure 4-3: Reactor concepts that Produce plutonium (a), and reactor concepts that consume plutonium (b).

development strategy makes sense?

4.3.1 Growing Plutonium Stocks

World plutonium levels, separated and in nuclear spent fuel, are large and growing rapidly. Trapp *et al.* [JTA+, 1993] estimate that there is presently 800 MT of plutonium from nuclear power reactors, and is increasing at a rate of 80 MT/year. As part of the effort by several countries to establish a plutonium-based fuel cycle, a portion of this civilian spent fuel has been processed to separate an amount of RGPu nearly equal that of WGPu. The OTA estimates that 100 MT of RGPu has been separated for this purpose [OTA, 1993]. Other estimates predict as much as 150 MT have been separated [IAE, 1992]. In addition, stockpiles of nuclear warheads in the US and FSU contain approximately 250 tons separated of WGPu [Perlman, 1992]. Only a portion of this WGPu is slated for disposition, as discussed in Chapter 1.

The rate of plutonium generation is dependent on the type of nuclear fuel strategy that is used, and in turn, is dependent on the reactor technology in production. In terms of plutonium production, reactors can be placed into one of two categories: net Producers, and net Consumers. Figure 4-3 represents these two cases. All reactors can be thought of as having a plutonium consumption rate, A, and a certain plutonium production rate, B, each in units of MT/GWe/year. In the case of a Producer, labeled P in the figure, the consumption rate of plutonium fueled from outside sources is offset by the production of new plutonium during the fission process from uranium, therefore, the production term is greater than the consumption term, A > B. For the Consumer, the opposite is true. The rate of production is less than the consumption rate, A > B.

The current LWRs are a Producer technology because they burn an all-uranium fuel, i.e.,

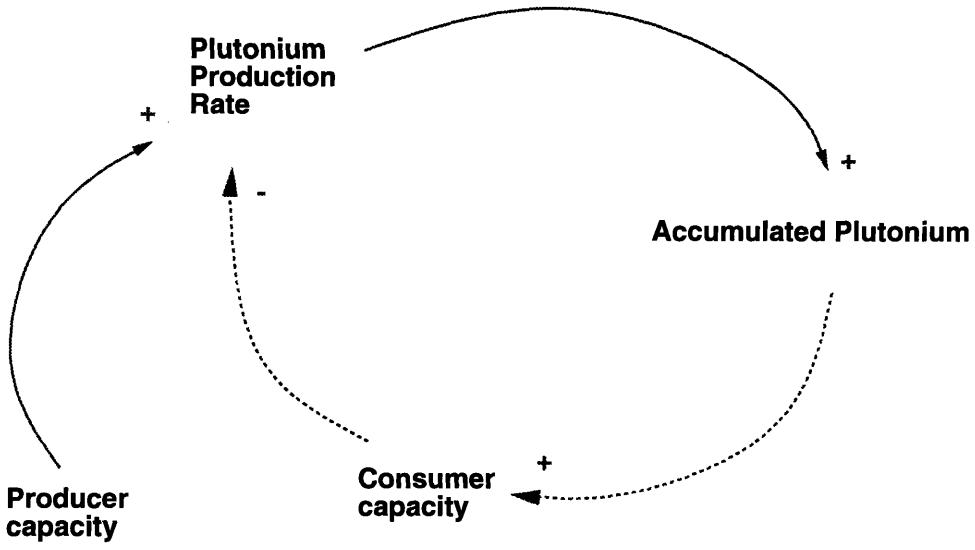

Figure 4-4: Plutonium accumulation resulting from net users and producers of plutonium.

A=0 and B is maximized. This represents the highest production rate of all reactor concepts, with nearly 40% of the original uranium fuel converted into plutonium. Considering MOX fuel strategies, plutonium is fed into the system (A > 0), however, since uranium is also present in the fuel assemblies, new plutonium is also produced (B > 0). Whether this system is a Producer or Consumer depends on the ratio of uranium-plutonium fuels, and the neutronics of the reactor design. The LMR reactor, for example, operates in the fast neutron energy spectrum, and can be designed to produce more plutonium than is fed in as MOX fuel. A variation of this concept has been proposed to consume the WGPu.

As the fuel strategy shifts to an all-plutonium fuel, the production term, B, becomes zero. The HTGR is one example of this case. This concept is able to achieve high burnups because a large excess reactivity designed into the system. The reactor concepts that display the highest plutonium consumption term is the ABC and MSR reactors. These concepts have a full-plutonium core, but also reprocess the fuel to achieve an almost complete burnup of the plutonium.

Knowing the plutonium generating character of each technology type, consider a simple model of world plutonium accumulation, as shown in Figure 4-4. This figure illustrates the cause and effect relationship between accumulated plutonium, and the type and amount of installed capacity of each type of reactor.

First, consider the installed Producer capacity. A net increase in Producer capacity

causes the Plutonium Production Rate to increase, which is indicated by the "+" sign. An increase in the Net Plutonium Production Rate then positively affects the Accumulated Plutonium. This describes the extent of the present nuclear fuel cycle. The total inventory of plutonium is linearly related to the total installed LWR capacity.

Now, consider the case where there is an installed base of Consumer capacity. In the situation, the Plutonium Production Rate is a function of installed capacity for both types of reactors. As the relative amount of Consumer capacity increases, the Plutonium Production Rate will decrease, which in turn will slow the accumulation of plutonium. Given this model, the level of accumulated plutonium cannot be reduced until the Net Production Rate is less than zero, i.e., the installed Consumer capacity is great enough to offset and exceed the Producer plutonium production rate.

Finally, consider the link between the installed Consumer capacity and the Accumulated Plutonium. As the amount of plutonium is reduced, the "fuel" available for the plutonium burning technology is reduced. This will force the installed Consumer capacity to decrease.

4.3.2 Simulating the Effect of New Reactor Technology on Plutonium Inventory

With this framework, a mathematical model was constructed to test the effect of various reactor types on the level of accumulated plutonium. A similar, but independent, study was conducted by Trapp [JDT, 1992]. The model begins with an initial installed capacity of LWRs, and assume constant nuclear capacity demand. Assuming a constant reactor decommissioning rate for all capacity in the system, as the LWR capacity is reduced by decommissioning, they are replaced with the plutonium-burning Consumer technology. Since the Consumer capacity technology is dependent on the amount of plutonium available, eventually, because the level of plutonium is reduced, the percentage of replacement capacity that burns plutonium is reduced. Eventually, equilibrium is reached, and the amount of plutonium burning capacity is just enough to burn what is generated by the all-uranium LWRs. Figure 4-5 shows an example where the LWR technology is replace with a once-through MOX Consumer reactor technology. As shown in the figure, as the LWR technology is decommissioned, the new MOX burning reactors replace the old, and the net electrical capacity remains constant. The control variables for the simulation include: (1) the percent of Consumer capacity installed, and (2), the unit consumption rate (MT/MWe/year) of the

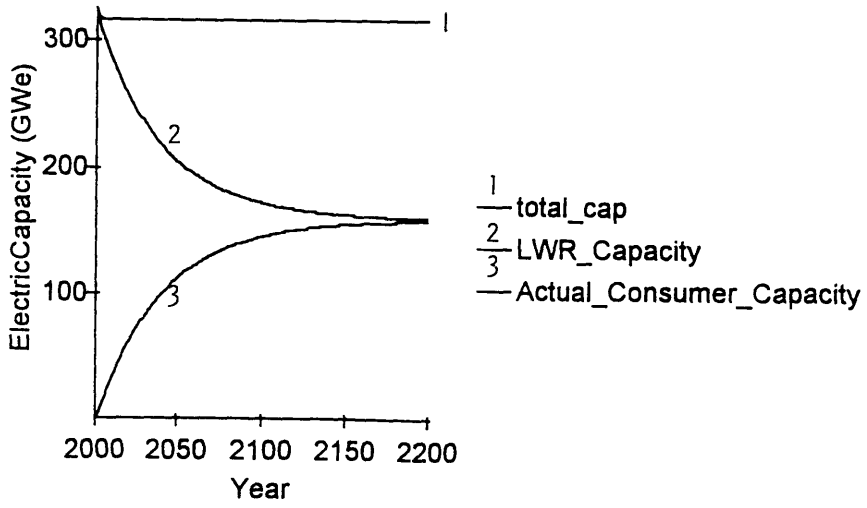

Figure 4-5: Introduction of Consumer reactors into installed capacity base.

Consumer technology. The following general assumptions were made:

- The initial stock of plutonium is 800 MT of RGPu plus 150MT of WGPu.
- There are only two types of reactors in the system, LWRs and the Consumer technology.
- 100% of presently installed nuclear capacity consists of LWRs.
- The demand for nuclear capacity is at a constant 325GWe [IAE, 1992]. Thus, for every GWe that is decommissioned, new capacity (new LWRs or Consumers) replaces the old so that the net capacity is constant.
- The average lifetime of all reactors is 40 years.
- Initially, 50% decommissioned reactor capacity is replaced by plutonium burning technology. This assumption forms an aggressive strategy for introducing Consumer capacity. However, the percent of Consumer is directly related with the amount of plutonium available in store. This means that as the stock of plutonium decreases, so too does the ratio of new Consumer capacity to new LWR capacity.

Table 4.1 lists the net plutonium generation rate for several technologies [MT/GWe/yr]. All except one are Consumer reactors. The ABC systems data was derived from an ex-

Concept	*Net Plutonium Generation*
(-)	MT/GWe/year
LWR(33,000MWD/MT)	0.30
LMR(converter)	-0.25
LWR(1 recycle)	-0.35
LWR(2 recycle)	-0.56
ABC	-1.20

Table 4.1: MT of plutonium generated per GWe capacity in one year of operation

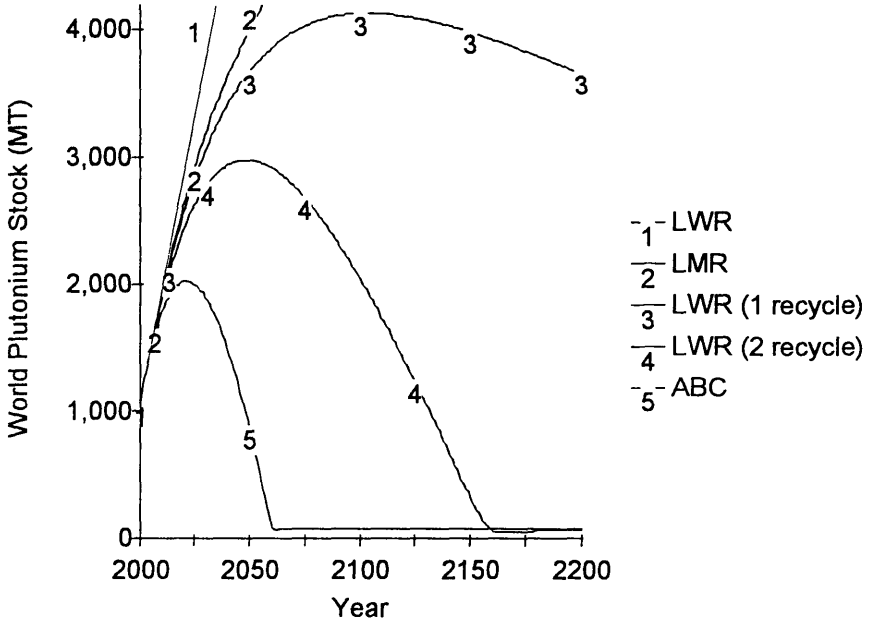

Figure 4-6: World Plutonium Inventory as a function of "Consumer" reactor technology

ample found in Arthur*et al.* [ADL, 1992]. The LWR and LMR data taken from Trapp [Trapp, 1993].

The model was simulated for the years between 2000 and 2200 and the results of are shown in Figure 4-6. As a baseline, the first simulation was run without introducing Consumer capacity. Instead, all decommissioned LWRs capacity was replaced with similar LWRs. In this case, as shown in the graph, the plutonium stock continues to rise at constant rate, curve 1.

Next, a LMR is introduced into the system to replace the decommissioned LWRs which has a plutonium consumption rate of .25 MT/GWe/year, curve 2. As the new technology begins to dominate the system, there is a marked reduction of the rate of plutonium accumulation, i.e. the slope of the curve is reduced. However, since the unit consumptions is low, there is little effect on the overall plutonium stock, at least in the time scales considered.

It is not until the introduction of the once-through MOX LWR reactor, with a consumption rate of .35 MT/GWe/year, that significant changes are seen. As indicated by curve 3, the plutonium level increases similarly to the first two cases until about the year 2100. At this point, because the unit consumption rate of the once-through MOX LWR times its corresponding capacity is equal to the generation rate of the remaining LWR capacity, the slope of the plutonium stock curve is zero. Then as the plutonium burning reactors continue to increase relative to the standard LWRs, the net plutonium generation rate is negative and the plutonium level begins to decrease. This behavior is also seen with the MOX scenario with two recycles, but with a smaller overall inventory and shorter time minimum plutonium inventory.

In the last case, an ABC system is introduced. As discussed previously, a full plutonium core coupled with continuous recycling allows the ABC to have a significant unit consumption rate, 1.2 MT/GWe/year. The results are shown in curve 5. In a similar fashion to the MOX recycle concept, but on a much smaller time scale, the rate of production shifts from net production to net consumption around 2020, and is rapidly consumed to near zero by 2065.

This model constitutes a simple representation of a complex and highly non-linear system, and, therefore, its intent is only to gain a general understanding of how plutonium burning technologies may affect the accumulation of plutonium by commercial nuclear systems. From the information above, the following conclusions can be made:

1. If a plutonium reduction strategy is deemed necessary, then technologies that consume plutonium will have to be developed to replace, or partially replace, existing LWRs.

2. As the unit consumption of plutonium is increased, as a function of reactor concept, the less time required to see significant changes in plutonium stocks.

3. Reducing the stocks of plutonium in the world, or even slowing its accumulation, is a long-term proposition.

4.4 Conclusions and Recommendations

As stated at the outset of this discussion, a technology concept must be evaluated with a clear view of the problem definition and the proposed solution. This is particularly critical

for large complex systems where the issues and goals that form the boundary of the problem area, are often not clearly defined. A case in point is the plutonium disposition problem, where the definition of the problem will determine what processing concept offers the best solution. For example, if the problem is defined with only WGPu in mind, the clear choice is to pursue concepts that are near-term, and able to offer relatively safe protection against diversion. However, as the previous section demonstrated, if the "problem box" is drawn around a larger area, to include the issue of RGPu, this conclusion is not as clear.

Therefore, to restate the previous conclusion, the WGPu disposition problem that is now being studied in the US and Russia, is likely to demand a near-term, mature technology, although it is not clear whether a fission-based or non-fission-based concept will emerge as the victor. The decision will likely be made on an economic basis, given that several near-term concepts seem capable of offering sufficient proliferation resistance. For the ABC concept, its relative technical immaturity makes it a less desirable option, notwithstanding its proposed ability to reduce the proliferation risk beyond that offered by the near-term technologies. However, within the larger context of all stockpiled plutonium (WGPu and RGPu) the functionality offered by the ABC becomes more attractive. This is true for other nuclear issues as well. For instance, if the destruction of actinides substantially reduces the risk of nuclear waste in a geologic repository, as some propose, then an ABC-like system would offer benefits beyond the capability of any near-term technology.

Given these points, there seems to be some benefit in carrying the ABC system further in its development, but to what extent remains unclear. Omberg *et al.* [OW, 1993] conclude, after their evaluation of the reactor concepts, that although the more mature technologies are more attractive to the immediate needs of plutonium disposition, at least one annihilation option should be investigated. Right now, the ABC concept is at the stage in development where the components need to be developed individually prior to dealing with system integration issues. Since many of these components have direct applicability to other related problems, e.g., the ABC on-line chemical processing technology could be used in the effort to clean up the U.S. defense wastes, then funding could be provided to develop common basic technologies rather than program specific subsystems. In this way, the ABC development effort would receive the support it needs, while the potential benefit from this investment is spread over a larger area.

Appendix A

Plutonium Inventory Systems Model

The plutonium inventory model was developed to test the effect of introducing various reactor technologies on the world's accumulation of plutonium, and consists of a simple mathematical representation of the generation and destruction of plutonium. Figures A-1, A-2,A-3, and A-4 show the model structure by sector.

Plutonium accumulation is modeled as a simple "stock-and-flow" with the level of plutonium dependent on the rate of plutonium generation and the rate of transmutation. The plutonium generation rate is simply the product of the plutonium generation per unit capacity and the total installed capacity in the system. In a similar fashion, the rate of transmutation is defined as the product of Consumer capacity installed, i.e., reactors that are net plutonium burners, and the rate of plutonium consumption per capacity of the technology.

Given this structure, the plutonium level is constant only if the rate of generation is equal to the rate of consumption. In the base case, where only uranium reactors are installed, the level of plutonium increases at a constant rate. This assumes, of course, that demand for nuclear capacity remains constant. If the plutonium consumption rate is greater than what is generated, then the level of plutonium will decrease. The details of these various scenarios are discussed in Chapter 4.

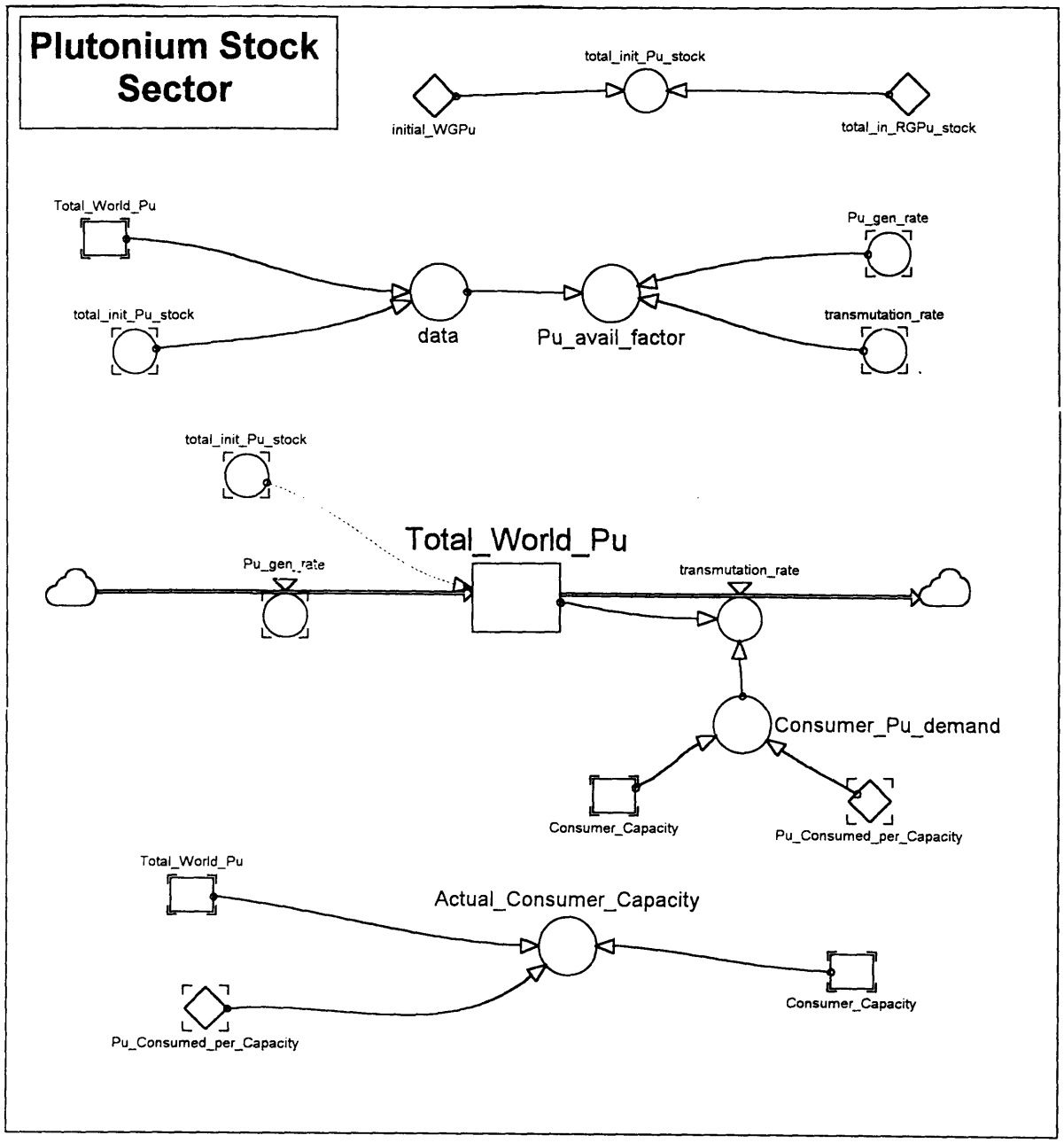

Figure A-1: Plutonium Stock Sector: Plutonium level is a function of Pu generation rate and plutonium transmutation rate.

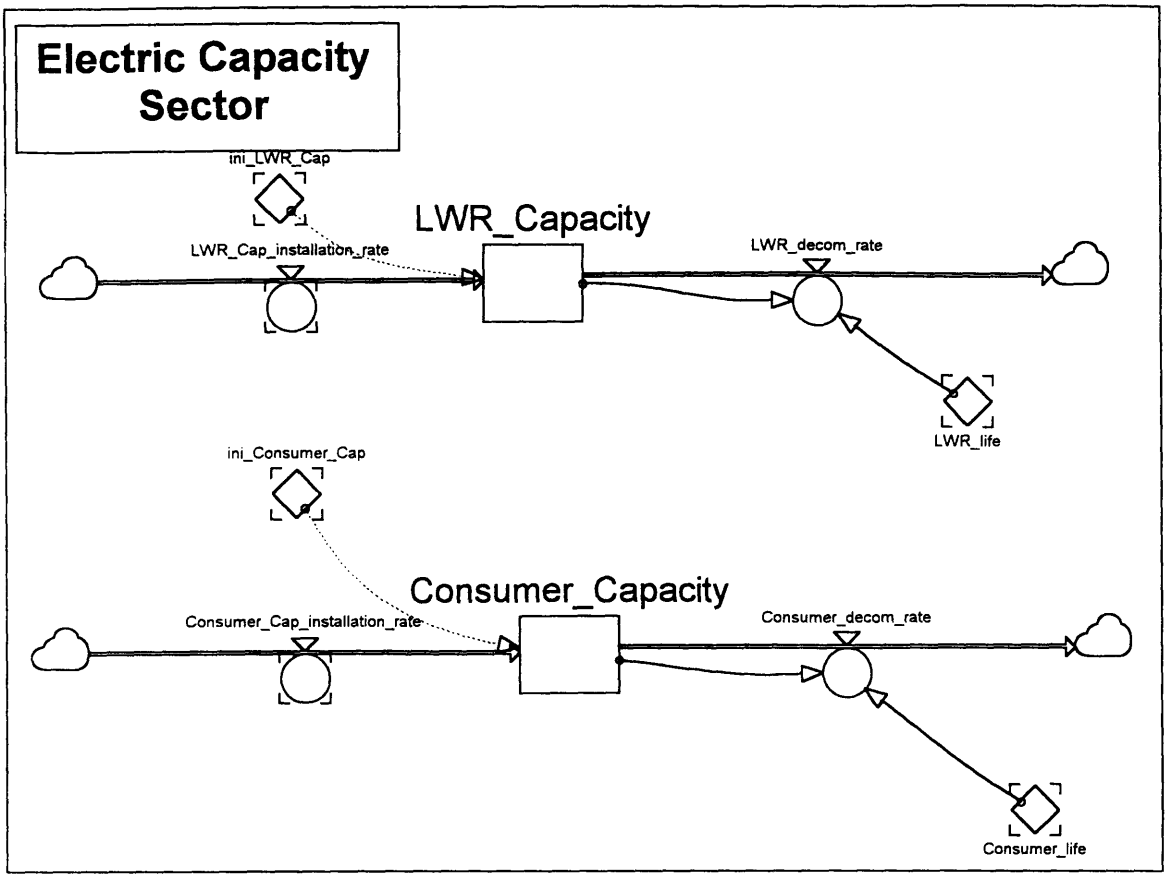

Figure A-2: Electric Capacity Sector: Electrical capacity of each technology is a function of the rate of installation and the decommissioning rate.

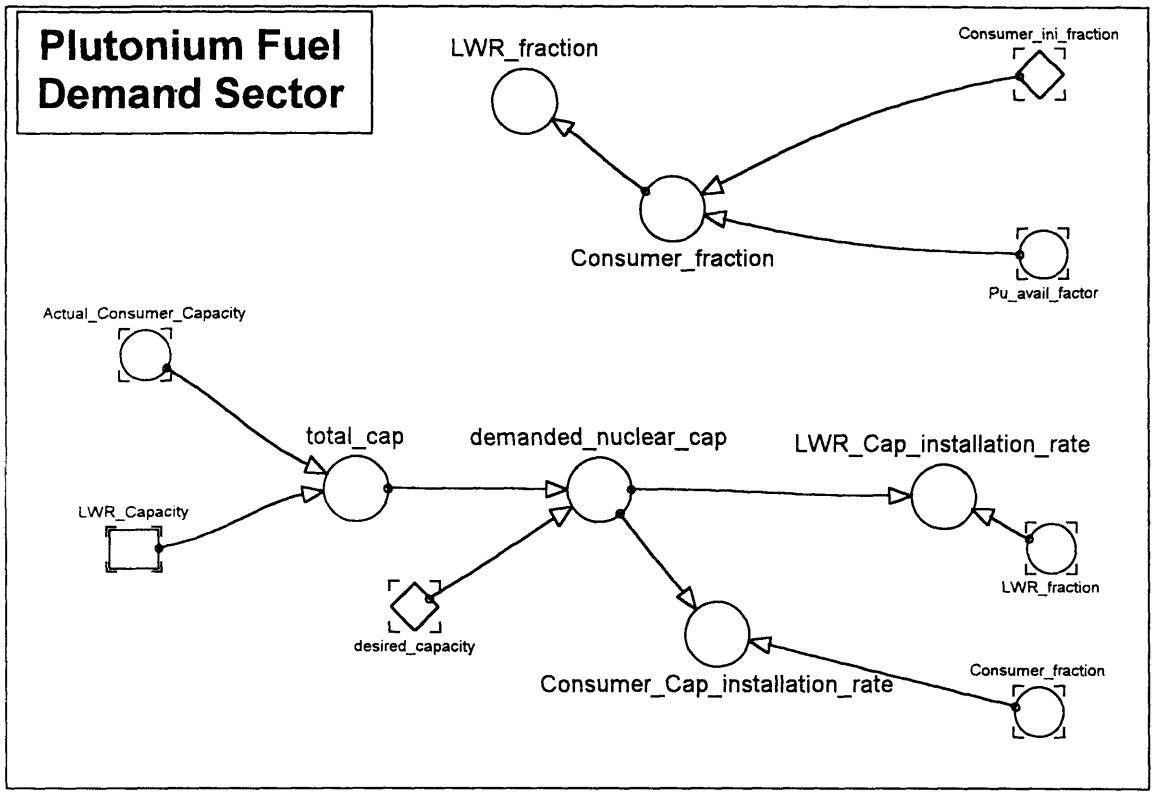

Figure A-3: Plutonium Fuel-Demand Sector: The required capacity to replace decommissioned reactors is distributed to each of the "competing" reactor types. The fraction to each is dependent on how aggressively the plutonium-burning technology is to be introduced.

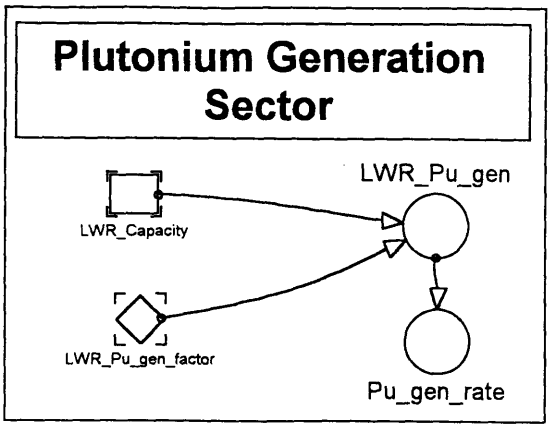

Figure A-4: Plutonium Generation Sector: It is assumed that the plutonium is generated only by the uranium-based reactor types. Although MOX reactors also generate plutonium, this model considers only their net affect.

Equation Listing

Actual_Consumer_Capacity
= IF(Total_World_Pu/Pu_Consumed_per_Capacity>Consumer_Capacity,Consumer_Capacity,Total_World_Pu/Pu_Consumed_per_Capacity)
Available Consumer capacity if the amount of plutonium is less than what is needed to fuel existing Consumer capacity.

Consumer_Cap_installation_rate
= demanded_nuclear_cap*Consumer_fraction
The amount of the total new capacity that is Consumer capacity.

Consumer_Capacity
INT ini_Consumer_Cap
-dt*(Consumer_decom_rate)
+dt*(Consumer_Cap_installation_rate)
Installed Consumer capacity.

Consumer_decom_rate
= IF(Consumer_Capacity<=0.0,0.0,Consumer_Capacity/Consumer_life)

Consumer_fraction
= Consumer_ini_fraction*Pu_avail_factor
This is the plutonium dependent fraction of new Consumer capacity that is installed. As the plutonium stock begins to diminish, the amount of new Consumer capacity that is installed begins to decrease.

Consumer_ini_fraction
= .5
Fraction of new capacity installed that is Consumer technology.

Consumer_life
= 40
Life time of Consumer plants is 40 years.

Consumer_Pu_demand
= Consumer_Capacity*Pu_Consumed_per_Capacity
Total amount of plutonium needed to operate installed Consumer capacity.

data
= GRAPH(Total_World_Pu/total_init_Pu_stock,0.00,0.50,[0,1,1,1,1,1,1"Min:0;Max:1"])

demanded_nuclear_cap
= IF(desired_capacity-total_cap<=0.0,0.0,desired_capacity-total_cap)
The amount of new nuclear capacity needed to maintain a constant total capacity.

desired_capacity
= 325
It is assumed that the total nuclear capacity is a constant 325 GWe.

ini_Consumer_Cap
= 0.00001
Initial Consumer Capacity is assumed to be zero.

ini_LWR_Cap
= 325
It is assumed that the total nuclear capacity is a constant 325 GWe. Initially, it is assumed that this capacity is totally LWR technology.

initial_WGPu
= 250
MT
This number is taken from a quote by Perlman (Perlman, 1992), and includes all plutonium stockpiled in warheads in the US and Russia.

LWR_Cap_installation_rate
= demanded_nuclear_cap*LWR_fraction
The amount of new capacity installed that is LWR technology.

LWR_Capacity
INIT ini_LWR_Cap
-dt*(LWR_decom_rate)
+dt*(LWR_Cap_installation_rate)
Installed LWR capacity.

LWR_decom_rate
= IF(LWR_Capacity<=0.0,0.0,LWR_Capacity/LWR_life)

LWR_fraction
= 1-Consumer_fraction

LWR_life
= 40
years
The assumed life time of all reactors is 40 years.

LWR_Pu_gen
= LWR_Capacity*LWR_Pu_gen_factor
Plutonium generation rate by present LWR technology.

LWR_Pu_gen_factor
= 0.3
This corresponds to the plutonium generated from a 33,000 MWD/MT average burnup LWR.

Pu_avail_factor
= IF(Pu_gen_rate>transmutation_rate,1,data)
Defines the effect of level of plutonium stocks with Consumer capacity.

Pu_Consumed_per_Capacity
= 1.2
Amount of net plutonium consumption using a particular Consumer technology.

Pu_gen_rate
= LWR_Pu_gen
It is assumed the total plutonium generated is the result of only the LWR technology.

total_cap
= LWR_Capacity+Actual_Consumer_Capacity
Total installed capacity in the world.

total_in_RGPu_stock
= 800
Total amount of plutonium generated to date from commercial reactors.

total_init_Pu_stock
= initial_WGPu+total_in_RGPu_stock
Total initial stockpile of plutonium.

Total_World_Pu
INIT total_init_Pu_stock
-dt*(transmutation_rate)
+dt*(Pu_gen_rate)
Total stock of putonium in the world (weapons-grade and reactor-grade)
transmutation_rate
= IF(Total_World_Pu>=Consumer_Pu_demand,Consumer_Pu_demand,Total_World_Pu)
MT/year
Total amount of plutonium burned by Consumer capacity per year.

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